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The first number of the Journal will appear early in 1956

CXXXVIII. *Non-Linear Regenerative Extraction of
Synchrocyclotron Beams*

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[Received September 12, 1955]

ABSTRACT

To extract a beam of a maximum energy from a synchrocyclotron, the deflector must be placed at the edge of the magnet, where the magnetic field falls off very rapidly and non-linearly with radius. This presents an analytical problem of coupled non-linear differential equations with periodic coefficients which is solved approximately. The results have been checked by electronic computation. It is concluded that a non-linear deflector can extract the beam with higher energy and perhaps greater theoretical efficiency than the linear one at present working at Liverpool. The new method is mechanically simpler than the old for it has no peeler, only a regenerator. The mathematics has interest apart from this application, for the difficulty of handling these coupled cyclotron equations is a major obstacle to the development of new particle accelerators of the fixed field type.

§ 1. INTRODUCTION

THE successful operation of the regenerative beam extractor in the Liverpool synchrocyclotron (Le Couteur 1951, 1953, Crewe and Le Couteur 1955) showed that the behaviour of the system was predictable and stable. However, the design was rather cautious and the high efficiency of operation entailed some sacrifice of energy; particles were extracted from 69 in. radius although the natural blow up of the internal beam occurs only at 71.5 in. Further analysis suggests that this limitation can be overcome but the system is so strongly non-linear that precise predictions require extensive numerical work.

§ 2. EQUATIONS OF MOTION

Let θ denote the azimuth and ρ and z the radial and vertical displacements of a particle from a synchronous orbit of radius r_0 , where the magnetic field is H_0 . At the position of the particle the vertical

* Communicated by the Authors.

magnetic field is $H=H(r, z)$ and, if the conservation of energy is used to express d^2/dt^2 as

$$d^2/dt^2 = \left(\frac{d\theta}{dt}\right)^2 \left(d^2/d\theta^2 - \frac{1}{r} \frac{dr}{d\theta} \frac{d}{d\theta}\right),$$

the radial Lorentz equation of motion becomes

$$\frac{d^2\rho}{d\theta^2} + \frac{r(rH - r_0H_0)}{r_0H_0} = \frac{1}{2r_0} \left\{ \left(\frac{d\rho}{d\theta}\right)^2 - \left(\frac{dz}{d\theta}\right)^2 \right\}, \quad . . . \quad (1)$$

with neglect of a term of third order in the velocities. This may be written

$$\frac{d^2\rho}{d\theta^2} + \rho + \frac{r_0}{H_0} (H - H_0) \left(1 + \frac{2\rho}{r_0}\right) + \frac{\rho^2 H}{r_0 H_0} = \frac{1}{2r_0} \left\{ \left(\frac{d\rho}{d\theta}\right)^2 - \left(\frac{dz}{d\theta}\right)^2 \right\} \quad (2)$$

or, if velocity ² terms are neglected and H expanded by Taylor's theorem, using $d^2H/dr^2 + d^2H/dz^2 = 0$,

$$\frac{d^2\rho}{d\theta^2} + \rho + \left(\frac{r}{H} \frac{\partial H}{\partial r}\right)_0 \rho \left(1 + \frac{2\rho}{r}\right) + \frac{1}{2} \left(\frac{r}{H} \frac{\partial^2 H}{\partial r^2}\right) (\rho^2 - z^2) + \frac{\rho^2}{r} = 0. \quad (3)$$

The vertical equation of motion is

$$\frac{d^2z}{d\theta^2} - \frac{1}{r} \frac{d\rho}{d\theta} \frac{dz}{d\theta} = \frac{r^2 H r}{r_0 H_0} \quad \quad (4)$$

or, neglecting velocity ² terms and using $\partial H r / \partial z = \partial H z / \partial r$,

$$\frac{d^2z}{d\theta^2} - z \frac{r_0}{H_0} \frac{\partial H(r)}{\partial r} \left(1 + \frac{\rho}{r_0}\right)^2 = 0. \quad \quad (5)$$

Further expansion yields the second order equations

$$\frac{d^2z}{d\theta^2} - z \left(\frac{r}{H} \frac{\partial H}{\partial r}\right)_0 - \rho z \left\{ \frac{r}{H} \frac{\partial^2 H}{\partial r^2} + \frac{2}{H} \frac{\partial H}{\partial r} \right\}_0 = 0. \quad \quad (6)$$

These forms of the equations of motion show that the radial motion depends on $(H - H_0)/\rho$ and the vertical on $\partial H / \partial r$; these quantities are significantly different in the present application.

Figure 1 shows a graph of the field H at the synchrocyclotron median plane and of $n = -(r/H)(\partial H / \partial r)$ and Hr . Let us consider deflector fields which start to act at $r_0 = 71$ in., so allowing normal acceleration to continue almost to the natural blow-up point. The analytical problem is clarified if the foregoing equations of motion are written explicitly as

$$\frac{d^2\rho}{d\theta^2} + \Omega^2 \rho = 0, \quad \frac{d^2z}{d\theta^2} + \omega^2 z = 0 \quad \text{for } \rho < 0, \quad . . . \quad (7)$$

$$\frac{d^2\rho}{d\theta^2} + \Omega^2 \rho (1 - 0.152\rho) + 0.13z^2 = 0, \quad \frac{d^2z}{d\theta^2} + (\omega^2 + 0.26\rho)z = 0$$

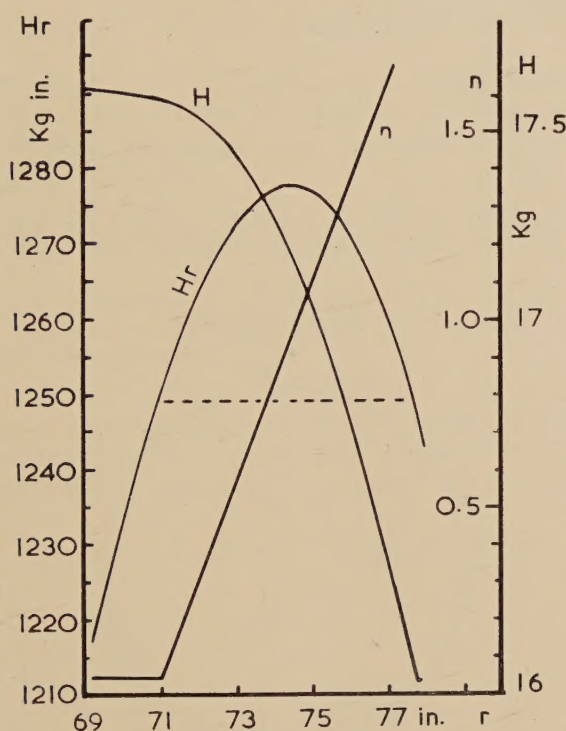
$$\text{for } \rho > 0, \quad . . . \quad (8)$$

where $\Omega^2 = 0.955 = 1 - n$ and $\omega^2 = 0.045 = n$ define the frequencies of radial and vertical oscillation in the linear region $\rho < 0$.

On the median plane the radial restoring force vanishes when $0.152\rho = 1$ or $\rho = 6.6$ in. This can be seen directly from fig. 1, for the values of

Hr at 71 in. and 77.6 in. are equal, so a particle with energy appropriate to 71 in. can also circulate on a metastable orbit at 77.6 in. and would have to be deflected beyond this radius in order to spiral out from the magnet without a magnetic channel. Such a large deflection does not seem to be advisable, so the problem is to swing the particles into a magnetic channel placed at about 74 in.

Fig. 1

Radial variation of H , Hr and $n = -(r/H)(\partial H/\partial r)$.

The deflector field H_q , of angular width θ_q at the regenerator vanishes at r_0 and increases outwards. According to (2) and (5) the regenerator produces impulses

$$\begin{aligned} \left(\frac{d\rho}{d\theta}\right)_{\text{out}} - \left(\frac{d\rho}{d\theta}\right)_{\text{in}} &= -\theta_q \frac{r_0}{H_0} H_q \left(1 + \frac{\rho}{r_0}\right)^2 \equiv -T_r \rho, \\ \left(\frac{dz}{d\theta}\right)_{\text{out}} - \left(\frac{dz}{d\theta}\right)_{\text{in}} &= \theta_q \frac{r_0}{H_0} \frac{\partial H_q}{\partial r} \left(1 + \frac{\rho}{r_0}\right)^2 z \equiv T_v z \quad \dots (9) \end{aligned}$$

and a peeler, if used, acts similarly with S replacing T . Evidently T_r and T_v are unequal if H varies nonlinearly with ρ ; it will be shown that this feature makes it possible to combine vertical stability with large radial deflections.

The equations formulated above are too complicated for exact solution and an adequate numerical survey of the problem would be a very large task, even for an electronic computer. So an approximate analysis of the strongly non-linear case has been used to determine suitable deflector fields and the expected performance was checked by a reasonable amount of orbit computation.

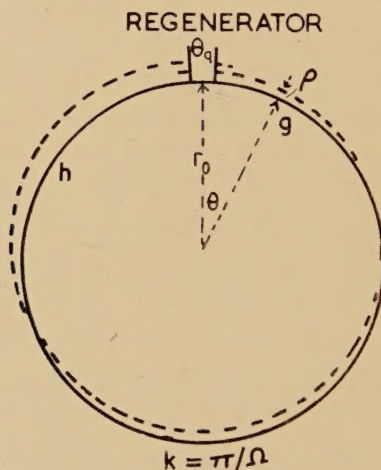
§ 3. RADIAL MOTION

The plot of H against r is approximated by two straight lines joining at $r_0 = 71$ in.

Then the radial motion of a particle with $\rho < 0$ is a simple harmonic motion with frequency Ω , defined by (7), and for $\rho > 0$, is a simple harmonic motion with a lower frequency Ω' . Encounters with the regenerator all take place with $\rho > 0$.

Consider the radial motion of a particle which crosses the radius r_0 at azimuth g after the regenerator, moving inwards as in fig. 2. After

Fig. 2



The dashed line indicates the path of a particle deflected by the regenerator from the synchronous orbit of radius r_0 .

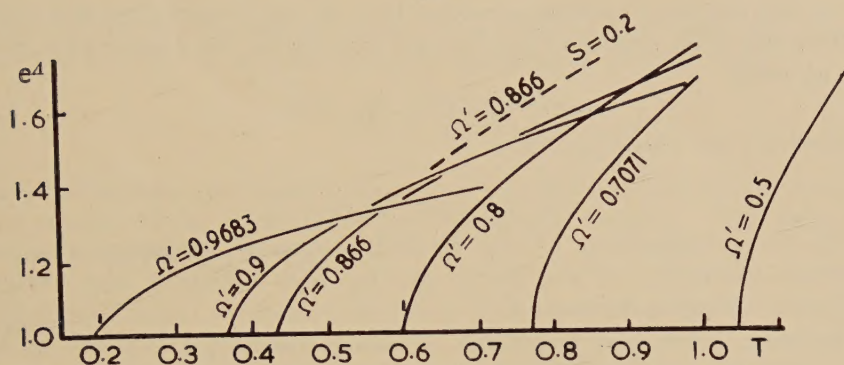
an angle $k = \pi/\Omega \simeq \pi$ the particle again crosses r_0 moving outwards with its original radial velocity reversed. This is exactly the effect which would be produced by traversing an angle $k' = \pi/\Omega'$ through a region of characteristic radial frequency Ω' . After a further angle h the particle re-enters the regenerator. Thus the regenerative radial motion for $\rho > 0$ may be calculated using the radial frequency Ω' everywhere, if the gap $f = g + \pi/\Omega + h$ is replaced by

$$f' = g + \pi/\Omega' + h = f + \pi/\Omega' - \pi/\Omega. \quad (10)$$

Accordingly the radial gain factor e^A may be calculated from (14) of 1951. Results are shown in fig. 3 for $n = 0.0625$, $\Omega = 0.9683$ and various

values of Ω' . The broken curve shows that a peeler of strength $S=0.2$ placed 60° before the regenerator, does not much increase the radial gain and further calculation shows that it does not improve the vertical

Fig. 3

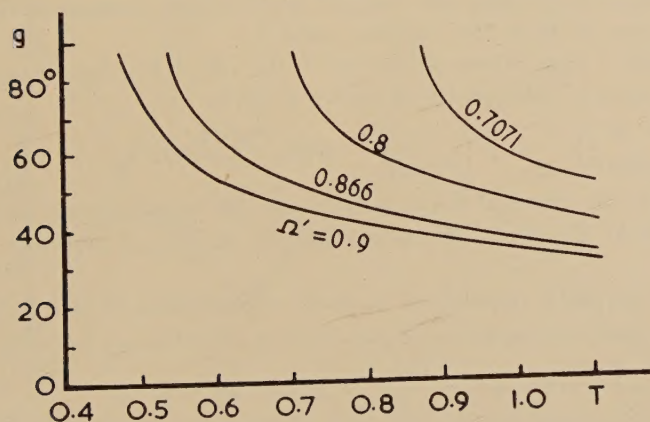


Radial gain e^4 as a function of regenerator strength T_r for $\Omega=0.9683$ and several values of Ω' . The broken curve shows effect of a peeler placed 60° before the regenerator.

focussing either. So it is best to work with a regenerator only, relying on the natural decrease of H in the region outside r_0 for the peeler effect. In this case formulae become very simple, (14) of 1951 reduces to

$$\cosh A = \cos \Omega' f' - \frac{T}{2\Omega'} \sin \Omega' f', \quad (11)$$

Fig. 4



Phase angle g with conditions as in fig. 3.

with $f' = 2\pi + \pi/\Omega' - \pi/\Omega$. If the azimuth θ is measured from the centre of the regenerator, after capture in the deflector the motion of a particle

leaving the regenerator for the m th time is

$$\rho = ce^{m\Delta} \sin(\phi + \Omega'\theta) \quad \text{for } \rho > 0, \quad \dots \quad (12)$$

with ϕ determined from 1953, eqn. (7), and 1951, eqn. (15) as

$$\cot \phi = (e^{\Delta} - \cos \Omega'f')/\sin \Omega'f'. \quad \dots \quad (13)$$

So in this motion ρ vanishes at azimuth $g = (\pi - \phi)/\Omega'$ and later at $g + \pi/\Omega$. Afterwards with ρ positive the particle approaches the regenerator again on an orbit

$$\rho = c'e^{m\Delta} \sin \Omega'(\theta - g - \pi/\Omega). \quad \dots \quad (14)$$

Values of g are plotted in fig. 4.

§ 4. VERTICAL MOTION

Careful consideration of the vertical motion is necessary to avoid vertical instability of the beam and for high efficiency the vertical expansion must be minimized.

Initially, when the amplitude of radial oscillation remains small, the third term in (6) may be neglected and the 1951 theory of constant n applies. Later when the particle is captured into the regenerative motion (12), (14), ρ is known as a function of θ so (5) or (6) become essentially linear differential equations with periodic coefficients. Then, in the approximation of § 3, the frequencies of vertical oscillation are

$$\left. \begin{aligned} \omega &= \sqrt{n} \quad \text{for } \rho < 0 \quad \text{in region } k, \\ \omega' &\quad \text{for } \rho > 0 \quad \text{in regions } g \text{ and } h. \end{aligned} \right\} \quad \dots \quad (15)$$

In the absence of deflector fields the vertical motion of a particle constrained to follow the radial path (5), (6), becomes unstable, because there is excessive disparity between ω' and ω when the amplitude of radial motion is large enough to make $\omega' \geq 0.55$. However, the regenerator can be designed to maintain vertical stability by compensating for the large decrease of H in the regions g and h .

As in (1951) eqn. (7) or Appendix A the effect on the vertical motion of one revolution through the deflector is given by

$$\begin{pmatrix} z \\ dz/d\theta \end{pmatrix}_{m+1} = A \begin{pmatrix} z \\ dz/d\theta \end{pmatrix}_m \quad \text{with } A = GQHK = \begin{vmatrix} \alpha & \beta \\ \gamma & \delta \end{vmatrix}, \quad (16)$$

where z_m is the vertical displacement at the m th passage through the radial node at the meet of g and k and G, H, K are matrices like D, F of 1951.

Vertical stability requires A to have eigenvalues of the form $e^{\pm i\lambda}$, so $\cos \lambda \equiv \frac{1}{2}(\alpha + \delta)$

$$\begin{aligned} &= \cos(\omega k + \omega'g + \omega'h) - \frac{(\omega' - \omega)^2}{2\omega\omega'} \sin \omega k \sin \omega'(g+h) \\ &+ \frac{T}{2\omega'} \left\{ \sin(\omega k + \omega'g + \omega'h) + \frac{(\omega' - \omega)^2}{2\omega\omega'} \sin \omega k \cos \omega'(g+h) \right. \\ &\left. + \frac{\omega'^2 - \omega^2}{2\omega\omega'} \sin \omega k \cos \omega'(h-g) \right\} \quad \dots \quad (17) \end{aligned}$$

should be numerically less than unity. When this condition is satisfied the deflector increases the initial vertical height h_0 of the beam by a factor

$$\frac{h_{\max}}{h_0} = (1 + \cos \epsilon / 1 - \cos \epsilon)^{1/2}. \quad (18)$$

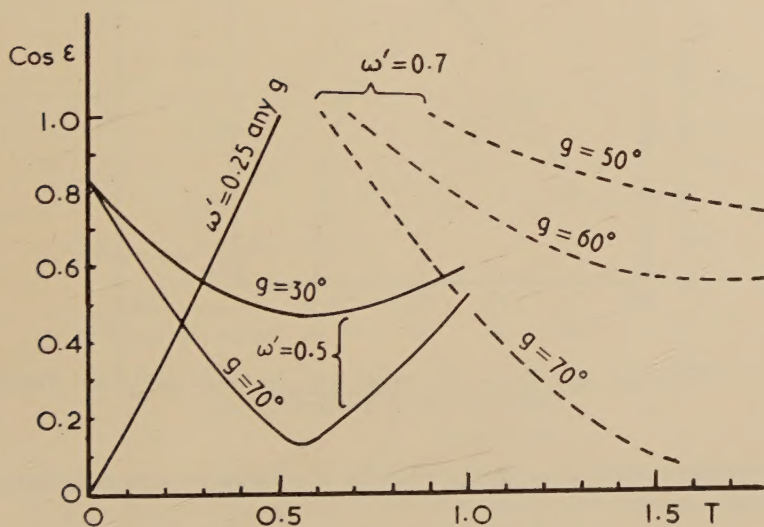
The vertical defocussing parameter $\cos \epsilon$ must be kept small and, for the motion in gap k is calculated, as in Appendix A of 1951, from

$$\sin \epsilon = \frac{2 \sin \lambda}{\beta \omega - \gamma / \omega}$$

and similar formulae apply to gaps g and h with ω' replacing ω ; $\cos \epsilon = 1$ corresponds to the limits $\cos \lambda = \pm 1$ of vertical stability. The values of β and γ defined by (16) give

$$\begin{aligned} \frac{1}{2}(\beta \omega - \gamma / \omega) = & \sin(\omega k + \omega' g + \omega' h) + \frac{(\omega' - \omega)^2}{2\omega\omega'} \cos \omega k \sin \omega'(g+h) \\ & - \frac{T}{2\omega'} \left\{ \cos(\omega k + \omega' g + \omega' h) + \frac{(\omega' - \omega)^2}{2\omega\omega'} \cos \omega k \cos \omega'(g+h) \right. \\ & \left. + \frac{\omega'^2 - \omega^2}{2\omega\omega'} \cos \omega k \cos \omega'(h-g) \right\} \end{aligned} \quad (19)$$

Fig. 5



Vertical defocussing, $\cos \epsilon$, for $\omega = 0.25$ and various values of ω' and g .

Putting $T=0$ in eqn. (17) shows that, in the absence of a regenerator, for $\omega = 0.25$ vertical instability sets in when $\omega' \geq 0.55$ so making $\cos \lambda < -1$. However the values of $\cos \epsilon$ plotted in fig. 5 show that vertical stability,

and even small vertical defocussing, can be maintained by proper choice of T . The values of $\cos \epsilon'$ are similar, but rather less as the vertical focussing forces are stronger in gaps g and h than in k .

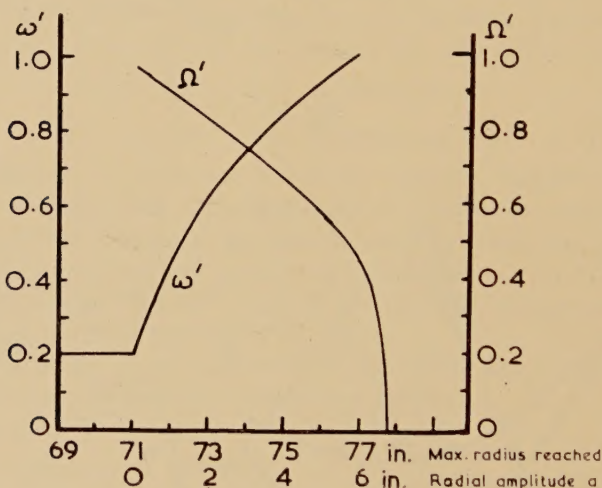
Similar analysis shows that in the difficult region of large ω' vertical defocussing is not reduced by use of a second deflector field, whether placed before or after the regenerator. So these complications are not discussed in detail.

§ 5. FREQUENCIES OF RADIAL AND VERTICAL MOTION

To make the best use of the results of § 4 the effective frequencies Ω' and ω' of radial and vertical motion in the region $\rho > 0$ must be carefully chosen.

For $\rho > 0$ and small z , the quarter period of the radial motion has been determined from (8) by means of an elliptic integral and, expressed as $\pi/2\Omega'$, determines the effective frequency Ω' as a function of the amplitude a of outward radial oscillation. These results are graphed in fig. 6.

Fig. 6



Effective frequencies Ω' and ω' of radial and vertical oscillation during an outward swing of amplitude a .

It does not seem necessary to give details of integration since the intuitive approximation

$$\text{quarter period} = \frac{\pi}{4} \left(\frac{1}{\Omega} + \frac{1}{\Omega \sqrt{1-0.152a}} \right) \quad \dots \quad (20)$$

is almost as good.

Equation (8) determines the average vertical oscillation frequency during an outward radial half cycle of amplitude a as approximately

$$\frac{1}{\pi} \int_0^\pi (\omega^2 + 0.26a \sin \theta)^{1/2} d\theta = 0.24\omega + 0.76(\omega^2 + 0.26a)^{1/2}, \quad (21)$$

which is also plotted in fig. 6.

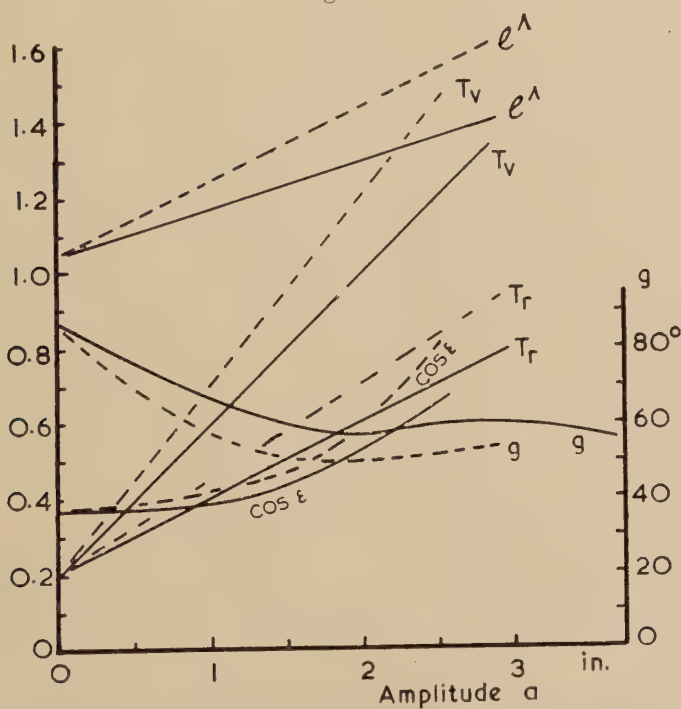
§ 6. CHOICE OF DEFLECTOR FIELDS

It is now possible to choose a suitable deflector field by combining the previous results.

At zero radial amplitude fig. 3 shows that $T_r > 0.2$ is required to initiate the regenerative motion.

At 2.5 in. radial amplitude, where $\omega' \simeq 0.7$, fig. 5 shows that to limit the vertical defocussing regenerator strength $T_v > 1$ and phase $g > 60^\circ$ are desirable, at this radius $\Omega' = 0.8$ and fig. 4 shows that the condition on g requires $T_r < 0.7$.

Fig. 7



Radial gain e^A , phase g and vertical defocussing $\cos \epsilon$ as functions of the amplitude a of outward radial oscillation for two regenerator fields. (a), full line $T=V=0.2$. (b), broken line $T=0.2, V=0.25$.

These various requirements are compatible. Consider a regenerator with

$$\theta_a \frac{r_0}{H_0} H_a = T\rho + V\rho^2; \quad (22)$$

according to definitions (9), then

$$T_r = T + \left(V + \frac{2T}{r_0} \right) \rho, \quad T_v = T + \left(2V + \frac{2T'}{r_0} \right) \rho. \quad (23)$$

The behaviour of a system with $T=V=0.2$ is shown in fig. 7(a), which indicates how radial gain e^A , phase g and vertical defocussing $\cos \epsilon$ vary with the amplitude of the outward radial motion. When the amplitude reaches 2.5 in. the radial gain is sufficient for entry into a thin-walled magnetic channel and the vertical defocussing is small enough for a high efficiency. After an initial decrease, the phase g does not vary much which implies that particles entering the channel should appear to diverge from a point source at azimuth $g+\pi$. Ω and incidentally validates the method of approximation used in this analysis.

Figure 7(b) shows results for a slightly stronger field which provides more radial gain at the cost of greater vertical defocussing.

§ 7. THE ELECTRONIC COMPUTER AT ROTHAMSTED

To check the approximate calculations a numerical solution of eqns. (7) and (8) was carried out on the electronic computer housed in the Statistical Department at Rothamsted. This machine is the '401' prototype model built by Elliott Brothers under contract from the National Research Development Corporation (Lipton 1955).

The 401 is a two address serial machine with a word length of 32 binary digits, one of which is a sign digit—this corresponds to about $9\frac{1}{2}$ decimal digits. The main store is a magnetic disc of 23 tracks, each track holding 128 words. All the usual operations and functions are available with the exception of division for which a sub-routine is used. Information is fed into the machine by 5-hole punched tape. Output is by electric typewriter or teleprinter punch.

§ 8. METHOD OF SOLUTION

A modified Runge-Kutta process was used (Wilkes, Wheeler and Gill 1951). Equations (7) and (8) were written as four simultaneous linear equations and to avoid overflow ρ , z , $\dot{z}$ were each scaled down by a factor of 10 and $\dot{\rho}$ by 100. (In the machine a number, t , lies in the range $(-1 \leq t < 1)$). Thus the equations to be solved were

$$\left. \begin{aligned} \dot{u} &= 10x \\ \dot{v} &= y \\ \dot{x} &= -0.0955u \\ \dot{y} &= -0.045v \end{aligned} \right\} \rho < 0, \quad \left. \begin{aligned} \dot{u} &= 10x \\ \dot{v} &= y \\ \dot{x} &= -0.955u(0.1 - 0.152u) - 0.13v^2 \\ \dot{y} &= -(0.045 + 2.6u)v \end{aligned} \right\} \rho > 0$$

where

$$10u = \rho, \quad 10v = z, \quad 10y = dz/d\theta, \quad 100x = d\rho/d\theta. \quad (24)$$

The scaled equations for the new values of $\dot{u}$ and $\dot{v}$ after regeneration (assuming $\rho > 0$) were

$$\begin{aligned}\dot{u}_{\text{out}} &= u_{\text{in}} - Tu - 10Vu^2 + 100Wu^3 \\ \dot{v}_{\text{out}} &= \dot{v}_{\text{in}} + Tv + 20Vuv - 300Wu^2v\end{aligned}\quad . \quad . \quad . \quad (25)$$

for a field given by

$$\theta_a \left(\frac{r_0}{H_0} \right) H_a = T\rho + V\rho^2 - W\rho^3. \quad . \quad . \quad . \quad (26)$$

The step length of integration was 4° . Results were printed out at values of θ of 328° and 360° during each revolution (the values at 360° were before regeneration). The information printed was the values of ρ , z , $dz/d\theta$, $d\rho/d\theta$, the maximum value of z reached so far and g , the value of θ at which ρ becomes negative, as in fig. 2.

§ 9. THE PROGRAMME

In order to apply the Runge-Kutta process three main routines are required. One of these is the basic integration routine which in turn uses an 'auxiliary' routine four times during each step. The purpose of the auxiliary is to calculate $\dot{u}$, $\dot{v}$, $\dot{x}$, and $\dot{y}$ from the values of the variables previously computed by the basic routine. Four such operations are required to advance the integration one step. In addition a 'master' routine coordinates the computations, keeps a count of the number of steps and prints the results when necessary.

A complication was introduced by the action of the regenerator. This difficulty was overcome by, in effect, restarting the computation at the end of each cycle of 90 steps: the values of ρ , z at this stage and the values of $d\rho/d\theta$, $dz/d\theta$ were used as new initial conditions. The master routine ensured that this operation occurred at the right value of θ , and took place only if $\rho > 0$.

Originally the programme was written so that the relevant defining eqns. (7) or (8) became operative as soon as ρ changed sign. This procedure was eventually altered and the range of negative ρ was crossed in one step in the following way.

If θ_n is the value of θ at which ρ becomes negative then it is permissible to jump immediately to $\theta_{n'}$, where

$$\theta_{n'} = \theta_n + 180^\circ / 0.955^{1/2} = \theta_n + 184^\circ$$

with

$$\begin{aligned}u_{n'} &= -u_n, & \dot{u}_{n'} &= -\dot{u}_n, \\ v_{n'} &= 0.7859v_n + 2.916\dot{v}_n, \\ v_{n'} &= -0.1311v_n + 0.7859\dot{v}_n.\end{aligned}$$

This modification considerably reduces the time taken to complete a whole revolution.

The Runge-Kutta has a truncation error in one step of order h^5 , where h is the step length, but rounding off errors accumulate at a rate inversely

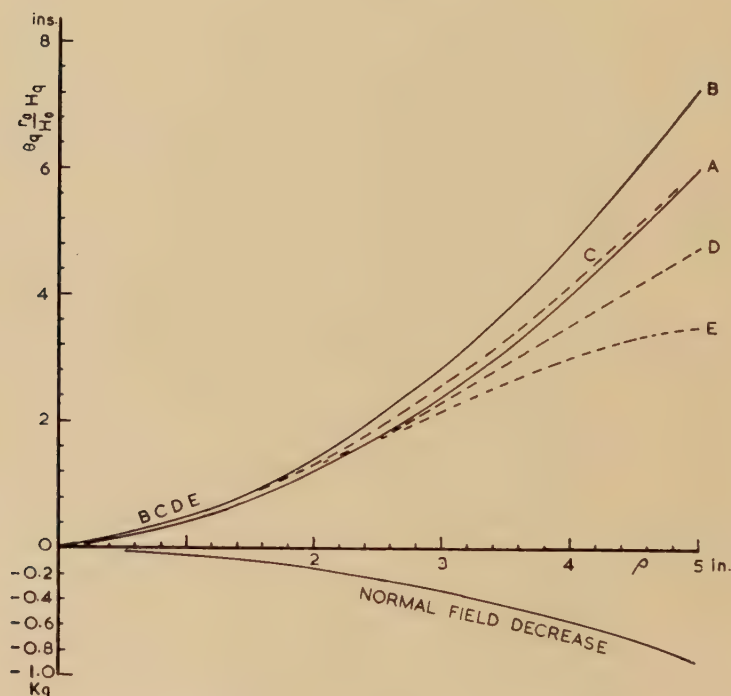
proportional to the number of binary digits held. The results are certainly correct to 3 decimal places.

Integration round a complete revolution with the modified programme described above took about 1 minute, including two rows of printing, and about 12 revolutions were obtained for each set of initial conditions. The solutions were computed over a period of four months, the work being carried out only during short intervals when the machine was not being used for its more usual statistical computations.

§ 10. RESULTS

Several starting values of the variables were used. One complete set of results served to indicate the effect for several different sets of initial condition, for each set of values printed at 360° in every cycle could be regarded as a new set of initial conditions. Thus we have the tracks of

Fig. 8



Regenerator field shapes H_q for cases A, B, C, D, E.

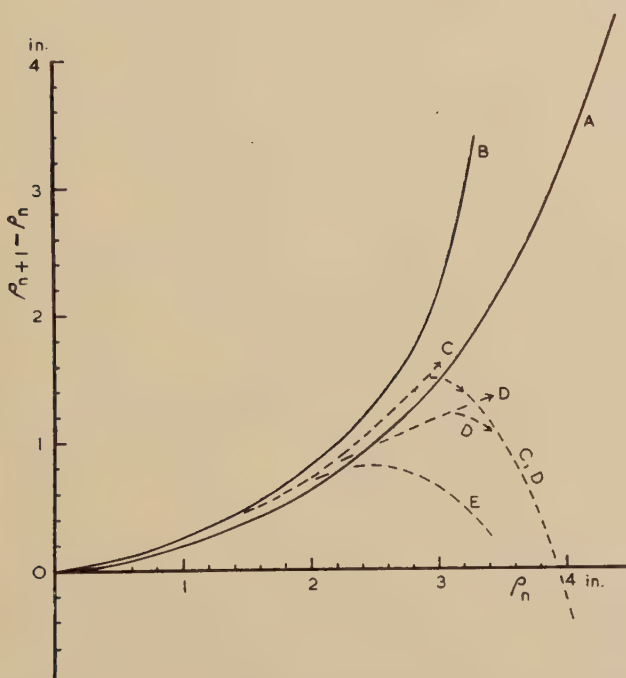
particles starting with initial values $\rho \approx 0.5$ in, $\dot{\rho} \approx 0$, $z = 0.5 \sin \alpha$, $\dot{z} = 0.5 \omega \cos \alpha$ for any phase α ; particles with smaller initial vertical displacements have smaller subsequent displacements approximately in proportion.

Table 1 lists the regenerator field shapes (26) which were tested, and for easy comparison these are shown in fig. 8 and their performance in figs. 9 and 10.

Table 1

Field	T	V	W
A	0.2	0.20	0
B	0.2	0.25	0
C	0.2	0.25	0.01
D	0.2	0.25	0.02
E	0.2	0.3	0.04

Fig. 9



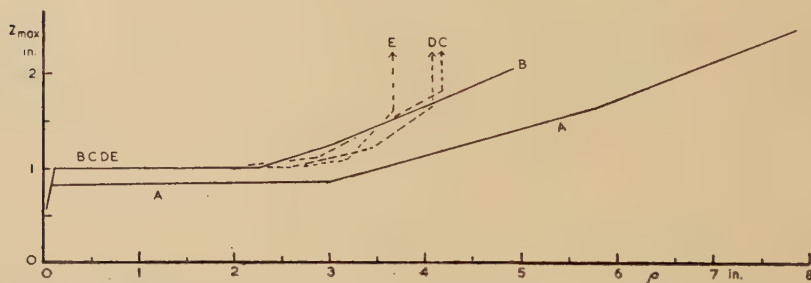
The increase, $\rho_{n+1} - \rho_n$, of successive radial displacements at 32° before the regenerator.

Figure 9 shows the radial separation $\rho_{n+1} - \rho_n$ of successive passages past azimuth 328° , that is 32° before the regenerator, together with the phase angle g of the motion. Figure 10 shows the maximum vertical displacement experienced anywhere by any particle with an initial amplitude of $\frac{1}{2}$ in., that is the height of the beam.

The vertical displacements at 328° , where an outlet channel might be placed, are usefully smaller than the maxima shown in fig. 10 because where ρ is large and positive the vertical focussing forces are very strong.

Clearly curves A and B give a satisfactory combination of limited vertical expansion and adequate radial separation, in reasonable quantitative agreement with the approximate results for these fields shown in fig. 7(a), (b). Fields C, D, E were used to study the effect of deviations from the shape indicated theoretically and are less satisfactory than A or B for they lead to dispersal of the beam when ρ exceeds 3 in. ;

Fig. 10



Maximum vertical displacement for any particle with less than 0.5 in. initial amplitude of vertical oscillation.

some particles go off vertically and retreat inwards while others continue outwards. For C at first sight this is surprising since it lies between A and B. The reason seems to be that for C, D, E the gradients dH_q/dr are less than for A and B and for $\rho > 3$ in. are too small to satisfy the vertical stability criteria established in § 5.

It is noteworthy that fields A and B produce radial displacements greater than 7 in., which carry particles beyond the point of free escape from the cyclotron.

§ 11. CONCLUSION

The theoretical analysis is easily adapted to any synchrocyclotron and shows that a non-linear regenerative deflector system can extract the beam with a higher energy than that now working at Liverpool. To get an intense external beam the particles must finally leave the machine through a magnetic channel, but since the deflector carries particles out to large radii the channel need not produce much field reduction. The resulting possibility of using a short magnetic channel of large aperture should lead to still greater efficiency than has already been achieved.

Whereas the linear apparatus requires two deflector fields, known as peeler and regenerator, the non-linear apparatus requires only one with a curvature in the sense which arises naturally when the field is produced by blocks of ions fixed in the synchrocyclotron.

The mathematics has some interest apart from the application to beam extraction, for the difficulty of handling the coupled non-linear equations (7) and (8) in the presence of similar periodic perturbations is at present one of the main obstacles to progress in the design of new particle accelerators of the fixed field type. The reasonable agreement found between results of a simple approximation and of extensive numerical computation suggests that these equations may be more tractable than is generally assumed.

These proposals were primarily intended for the Liverpool and C.E.R.N. synchrocyclotrons, but are now being applied to several others. The expected results have already been obtained at Chicago, by Dr. Crewe, using the deflector fields shown in fig. 7(a).

APPENDIX

As the precise characteristics of the deflected beam are important, table 2 gives particle coordinates at the last few successive passages past azimuth 328° for field A and various initial conditions. ρ vanishes at azimuth $g \pm 2^\circ$ and then at azimuth $g + 184 \pm 2^\circ$, which is the virtual source of particles swinging outwards.

Table 2

Revolution	$\rho(\text{in.})$	$z(\text{in.})$	$dz/d\theta$	$d\rho/d\theta$	$z_{\max}$	$g(^\circ)$
initial	0.5	0.5	0	0		
10	2.57	0.53	-0.40	0.64	0.86	58
11	3.57	-0.77	0.62	1.11	-1.02	58
12	5.80	0.82	-1.53	2.48	1.58	58
13	>10					
initial	0.5	0	0.125	0		
10	2.17	0.39	-0.07	0.51	0.72	58
11	2.96	0.18	-0.24	0.90	0.72	62
12	4.41	-0.55	0.67	1.69	0.86	58
13	8.40	0.82	-1.55	4.71	2.35	54
initial	0.5	0.2	0.1	0		
10	2.09	0.76	-0.30	0.38	0.86	58
11	2.89	-0.26	-0.00	0.79	0.86	58
12	4.36	-0.28	0.46	1.66	0.86	58
13	8.50	0.63	-1.21	4.94	1.82	54
initial	0.5	0	0.0125	0		
11	2.14	0.054	-0.020	0.63	0.07	62
12	2.88	-0.011	-0.006	0.96	0.07	62
13	4.33	-0.032	0.043	1.75	0.07	62
14	8.94	0.067	-0.079	5.62	0.16	54

ACKNOWLEDGMENTS

The numerical work was carried out on the electronic computer housed in the Statistical Department at Rothamsted. We are much indebted to Dr. F. Yates, F.R.S., the head of the Department and to the National Research Development Corporation for permission to use the computer on this problem. The cost of the computation was met by a grant from the C.E.R.N. synchrocyclotron division.

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CXXXIX. *The Nuclear Scattering of Electrons and Positrons at 10 mev*

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SUMMARY

Measurements have been made on the single (large angle) and multiple coulomb scattering of electrons and positrons in nuclear emulsions. The results obtained for single scattering are in general agreement with the theoretical expectations, and appear to confirm the prediction of a difference between the nuclear scattering of electrons and positrons at 10 mev, although the data are insufficient to permit detailed comparison. The results for the multiple scattering show an unexplained difference between the two types of particle. The number of positrons observed to annihilate in flight is in agreement with the theoretical predictions of Dirac (1930).

§ 1. SINGLE SCATTERING

(i) *Introduction*

THE single or large angle scattering of relativistic electrons has been studied by several workers at energies less than 4 mev, by Lyman, Hanson and Scott (1951) at 15.7 mev, and by Hofstadter *et al.* (1954) at higher energies. In contrast with the widely varying results of earlier experiments the more recent work at the lower energies is in general agreement with theoretical predictions. Apart from the work of Fowler and Oppenheimer (1938) at about 11 mev, in which only a few deflections of over 14° were observed, the single scattering of positrons has been studied only at low energies (≤ 2 mev; Howatson and Atkinson 1951). The nuclear scattering of electrons and positrons with energies in the range from 4–15 mev provides a test of the validity of the relativistic scattering formula for point charges, but can give little information about the nuclear charge distribution. In the present experiment the single scattering cross sections at 10 mev for electrons and for positrons were examined under conditions which enabled a direct comparison to be made between them.

(ii) *Exposure of the Emulsions*

The Glasgow H.T. set was used to provide the 10 mev positrons and electrons by means of the nuclear reactions ${}^7\text{Li}(p, \gamma){}^8\text{Be}$ and ${}^7\text{Li}(d, p){}^8\text{Li}$.

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† Communicated by Professor P. I. Dee, F.R.S.

In the first case both types of particle were produced from the 17.6 mev gamma-rays by pair production in lead, and in the second case a large flux of electrons with a maximum energy of about 12 mev was obtained from the decay of the short-lived ^8Li nuclei. All the positrons were produced from the first reaction, and electrons were obtained by both methods. The greater yield from the $^7\text{Li}(d, p)^8\text{Li}$ reaction allowed much shorter times of exposure than were possible in the case of pair production. The particles produced in these reactions were analysed by a double focusing magnetic spectrometer which had a resolution rather better than 5% for the absolute value of the energy, and were then allowed to pass into 400 μ Ilford G5 emulsions. The magnetic field was measured at intervals during the exposure of each plate and the corresponding energies in all exposures were close to 10 mev. The final results have all been normalized to this energy.

(iii) Analysis

The measurements were made on Cooke M. 4000 microscopes using $\times 15$ eyepieces and $\times 45$ oil immersion objectives. The plane of the emulsion was inclined at $\sim 9^\circ$ to the beam of particles which extended over the central inch of each three-inch plate. Only tracks from the central portion of the beam were used for measurement since a check, by multiple scattering measurements, on the energies of particles distributed over the beam revealed a few rather low-energy particles on the fringe of the beam in one exposure. Apart from this limitation the acceptance criteria for a track were

- (a) that it should lie within 10° of the mean beam direction,
- (b) that it should have a minimum length of 500 μ for surface to surface tracks and of 700 μ for surface to glass tracks.

Suitable tracks were followed from their point of entry into the emulsion to a maximum length of 2000 μ . For all scattering angles having a horizontal projection greater than 10° measurements were made of the horizontal projection of the angle of scattering and of the dip angle of the primary and scattered tracks. From these measurements the spatial angle of scattering was calculated in each case. Track lengths were measured using an eyepiece scale and corrected for the effect of dip. The experimental results were corrected for effects due to

- (a) the rejection of scattering angles with horizontal projections $< 10^\circ$,
- (b) the rejection of tracks having lengths less than 500 or 700 microns.

(iv) Results

380 cm of electron track and 520 cm of positron track were scanned giving total numbers of deflections $> 25^\circ$ of 136 for electrons and 163 for positrons. The resulting probabilities for scattering in the emulsion, after the application of the correction factors mentioned above, are shown in table 1. The errors indicated are standard deviations,

Table 1 also gives the anticipated theoretical values for the probability of scattering as obtained from the calculations of Feshbach and others (Feshbach 1952, McKinley and Feshbach 1948, Acheson 1951) for electrons and Yadav (1952) for positrons. In obtaining the theoretical values we have evaluated the sum $\sum_i N_i \sigma_i(\theta_1 - \theta_2)$ where $\sigma_i(\theta_1 - \theta_2)$ is the scattering cross section of the i th element for the angular interval $(\theta_1 - \theta_2)$, N_i is the number of atoms of the i th element present per cm^3 of the emulsion, and where the sum extends over all the elements present in the emulsion. Since σ_i is proportional to z_i^2 , where z_i is the atomic number of the i th element, nearly all the scattering is due to the silver and bromine content of the emulsion. The effect of finite nuclear size has been included in the calculation of the cross sections by using the results of Acheson (1951) and of Elton and Parker (1953), but the effect is small ($\sim 7\%$), even for the highest angular interval.

Table 1. Probability of Scattering per cm of Track Length in the Emulsion

Angular interval		25°-35°	35°-50°	50°-180°
Electrons	Measured	0.275 ± 0.031	0.120 ± 0.020	0.110 ± 0.019
	Theoretical	0.280	0.150	0.102
Positrons	Measured	0.220 ± 0.024	0.115 ± 0.017	0.066 ± 0.013
	Theoretical	0.203	0.095	0.054

The statistical errors associated with the experimental data are too large to allow a detailed examination to be made of the differences in the scattering probabilities for electrons and positrons as a function of scattering angle. The results do, however, appear to confirm the theoretical prediction of a difference between the nuclear scattering of electrons and positrons at 10 mev due to the differences in the spin-orbital forces on the two types of particles, and also show that the theories of single coulomb scattering at this energy predict cross sections of the right magnitude.

§ 2. MULTIPLE SCATTERING

The multiple coulomb scattering of electrons and positrons in nuclear emulsions has been studied by several workers (Corson 1951, Gottstein *et al.* 1951, Bosley and Muirhead 1952, Husain 1955, Williams, private communication).

Direct comparisons between the multiple scattering of electrons and positrons with energies of about 2 mev in argon have been made by Groetzinger, Humphrey and Ribe (1952), and by Cusack and Stott (1955),

using a cloud chamber. A study of the scattering in emulsions of both types of particle under the same conditions has been made by Corson using particles of 196 mev and by Williams using particles of 1, 1.5 and 2.5 mev. The present work enables a direct comparison to be made between the scattering of electrons and positrons with an energy of 10 mev.

Earlier work in this laboratory with a magnet of poorer resolving power had indicated a difference between the multiple scattering of electrons and positrons. In the present work tracks were measured on the same plates as were used for the single scattering experiment, and the relative energies of the two types of particle should be correct to about 2%.

Scattering measurements using the coordinate technique developed by Fowler (1950) were made on about 9 cm of electron track and about 10 cm of positron track, and from these results values of the scattering constant K for the emulsion were calculated from

$$K = \langle \phi \rangle_{\text{CHORD}} \frac{p\beta c}{z} \left(\frac{100}{t} \right)^{1/2}$$

where $\langle \phi \rangle_{\text{CHORD}}$ is the mean angle of scattering measured by the coordinate method, p , βc are the momentum and velocity of the scattered particle, z is the ratio of its charge to that of the electron, and t is the cell size in microns. K is expressed in the usual units of degrees $\times$ Mev $\times$ (100 μ) $^{-1/2}$.

The measured values of K were corrected for the effects of spurious scattering due to irregularities in the microscope movement and to observer errors ('noise'), for the inclination of the tracks to the coordinate axes used for measurement, and for distortion of the emulsion. Except for the very small cell sizes where the effect of 'noise' is appreciable ('noise' correction for 25 μ cell is $\sim 10\%$), all these corrections are small.

The corrected values of K are plotted in fig. 1(a) as a function of cell size. A value of the scattering constant less liable to be affected by variations in the small number of large deflections in the distribution is obtained by omitting angles greater than four times the median value. These 'cut' scattering constants K_c are plotted in fig. 1(b).

In table 2 the results obtained from the present work are compared with the theoretical predictions and with results obtained by other workers. The theoretical results quoted here are those of Molière (1948), and take no account of the spin of the particles. The magnitude of the difference in scattering to be expected from this effect has been calculated by Mohr and Tassie (1954); their results predict a difference in scattering between positrons and electrons of less than 1% under the conditions of the present experiment. The results quoted in the table do not include those of Williams (*loc. cit.*) for the scattering of 1, 1.5 and 2.5 mev electrons and positrons in nuclear emulsions, which are presented in a forthcoming paper, where a difference is also indicated. The results

from the cloud chamber measurements are not included in the table, since they do not yield values of a scattering constant directly comparable with those obtained using nuclear emulsions. The measurements of Groetzinger, Humphrey and Ribe (*loc. cit.*), at 1.5–2.5 mev show a difference of about 10% between the scattering of electrons and positrons compared with a theoretical prediction $\sim 1\%$. Cusack and Stott (*loc. cit.*), at 0.4 mev find no significant difference between the scattering of the two types of particle.

Fig. 1

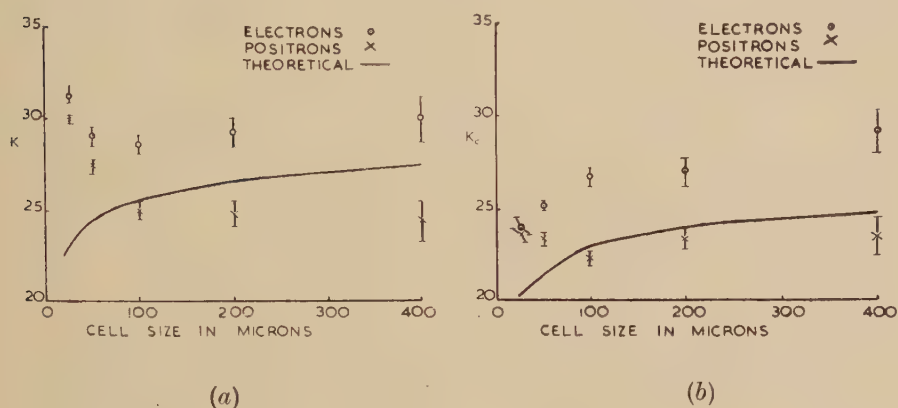


Table 2. Values of the Scattering Constant K for 100 μ Cell Size

Author	Positrons	Electrons
Present work, 10 mev	24.9 ± 0.5	28.6 ± 0.5
Gottstein <i>et al.</i> , 105 mev	25.8 ± 0.6	—
Corson, 196 mev	25.0 ± 0.8	27.3 ± 0.8
Bosley and Muirhead, 9–19 mev	—	26.5 ± 1.2
Husain, 6.6 and 11.5 mev	—	23.6 ± 1.2
Theoretical (Molière)	25.5	25.5

§ 3. ANNIHILATION OF POSITRONS IN FLIGHT

The positron plates showed 18 examples of particles which appeared to annihilate in flight. The number calculated from the theory of Dirac (1930) is 22.

ACKNOWLEDGMENTS

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CXL. *Nuclear Orientation of Cerium 141*

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ABSTRACT

^{141}Ce has been aligned by the method of magnetic hfs (alignment) in a crystal of cerium ethyl sulphate. From measurements of the angular distribution and the plane of polarization of the γ -radiation it is concluded that the γ -ray transition is principally M1 with a small admixture of E2 ($\delta = +0.08 \pm 0.02$). Values of 0.75 ± 0.20 and 0.66 ± 0.16 nuclear magnetons are found for the nuclear moment of ^{141}Ce , depending on whether the β -transition involves a spin change of 1 or 0.

§ 1. INTRODUCTION

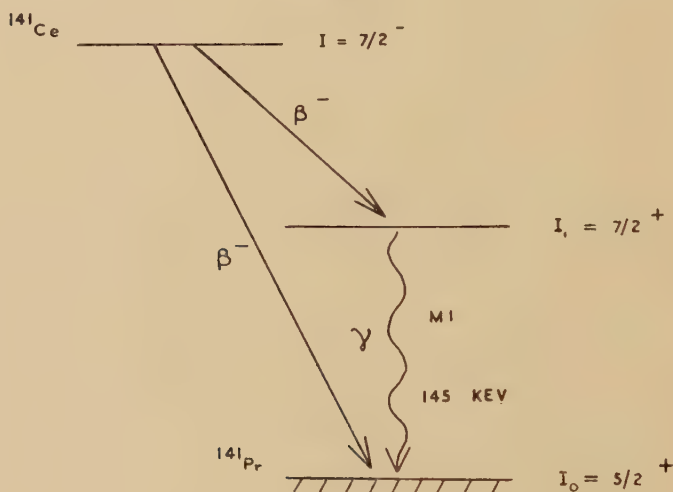
THE method of nuclear orientation applied to the investigation of nuclear properties by observing the directional and polarization effects of the γ -radiation from oriented radioactive nuclei has been described in earlier papers (Bleaney *et al.* 1954, Bishop *et al.* 1955 a). It was indicated in these that for a given cascade of β - and γ -ray transitions and for a given assignment of spins to the associated levels the anisotropy of a particular γ -ray depends on the multipole order of the transition, on the character of previous transitions and on the original degree of orientation. It was also pointed out that quite small coherent admixtures of other multipoles in the γ -ray transition can produce an angular distribution radically different from that of the pure transition. At the same time this admixture will alter the degree of plane polarization of the radiation but to a different extent. For instance, it is possible to destroy the γ -ray anisotropy completely and yet leave a large polarization. Conversely it is possible from simultaneous measurements of the γ -ray anisotropy and polarization to determine the mixing ratio in mixed γ -ray transitions. This method of studying mixed transitions has now been applied to the isotope ^{141}Ce which was oriented using the method of magnetic hfs (alignment); this isotope was first aligned by Ambler, Hudson and Temmer (1955). The present experiment differs from that of Ambler *et al.* in two respects. Firstly a crystal was used which gave nuclear alignment along an axis in contrast to alignment in a plane. Secondly the polarimeter was employed which showed that the γ -ray contained a mixture of multipole radiations.

* Communicated by the Authors.

§ 2. DECAY SCHEME OF ^{141}Ce

The principal features of the decay scheme of ^{141}Ce are well known and are shown in fig. 1 (Jones and Jenson 1955). The spin of the ground state of ^{141}Pr has been measured as $5/2$ and from its nuclear moment this is identified as a $d\ 5/2$ state on the nuclear shell model. The assignment of $f\ 7/2$ to the ground state of ^{141}Ce is made on the basis of comparative half-life values for the β -transitions which suggest that both are first forbidden with $i_\beta = 0$ or ± 1 . Internal conversion (Heath 1952) and lifetime measurements (de Waard and Gerholm 1955) on this γ -ray indicate that it is principally M1 and this appears to be confirmed by the nuclear orientation measurements of Ambler *et al.* These results can be most easily understood by an assignment of $g\ 7/2$ to the first excited state of ^{141}Pr (see fig. 1).

Fig. 1



The decay scheme of Cerium 141.

Temmer and Heydenburg (1954) have shown that the 145 keV state is weakly excited by Coulomb excitation of ^{141}Pr and we conclude that at least part of the γ -ray transition must be electric (presumably E2). By comparing their figures on the intensity of this radiation with that obtained in ^{181}Ta we can estimate the partial half-life for E2 decay as approximately 3×10^{-6} sec. Direct measurement of the half-life of this state for γ -ray emission by de Waard and Gerholm (1955) gives a value of $2.4 \pm 0.3 \times 10^{-9}$ sec and we therefore conclude that the ratio of the intensities E2/M1 is about 10^{-3} . Since the half-life estimate from Coulomb excitation is very approximate this value for the ratio of the intensities is only correct to within an order of magnitude.

§ 3. PREVIOUS NUCLEAR ORIENTATION RESULTS

The experiments of Ambler *et al.* were made using a crystal of cerium magnesium nitrate. In this crystal the cerium nuclei become oriented in such a way that the nuclear magnetic state with $I_z = \pm \frac{1}{2}$ is preferentially populated, i.e. the nuclei tend to align with their spins in a plane perpendicular to the z -axis. From the observed γ -ray angular distribution and its dependence on temperature these authors conclude that the transition is M1 and also that the spin of the 145 keV state is $7/2^+$. To determine the nuclear moment of ^{141}Ce it is necessary to know the hfs field for cerium in this crystal. The hfs for cerium has not been measured experimentally since no stable isotope with a moment different from zero exists. However, using calculated values for the hfs separation they deduced that the nuclear moment is 0.16 nuclear magnetons. This is smaller than that to be expected for a nucleus whose moment arises from a single $f 7/2$ neutron; other nuclei with $f 7/2$ neutron configurations have moments of about 1 nuclear magneton (Walchli 1953). This small value could arise from two possible causes. Firstly, there might be a reduction in the hfs field due to interaction between the cerium ions, not taken into account in the calculation. This could give rise to a smaller degree of orientation of ^{141}Ce and hence lead to a smaller value of nuclear moment. This is discussed by Ambler *et al.* Secondly, there might be a reduction in the γ -ray anisotropy either due to a perturbation of the first excited state of ^{141}Pr through interaction with extranuclear fields, or to the presence of a coherent admixture of E2 radiation in the M1 transition. It seems reasonable to suppose that in planar alignment, where the lower levels are relatively closer together, the nuclei would be more likely to be perturbed by extra-nuclear fields than in axial alignment.

§ 4. DESIGN OF EXPERIMENT

To avoid those difficulties which may be present in the cerium magnesium nitrate crystal we decided to align this nucleus in a crystal where axial alignment should be obtainable. This is a more favourable case for obtaining orientation as the splitting between the lowest levels is in general larger (Bleaney 1951). Because of lack of knowledge of the hyperfine structure field in cerium one must be guided by g -values in choosing the crystal. In cerium ethyl sulphate $g_{\parallel} = 3.9$, $g_{\perp} = 0.2$ (Bogle, Cooke and Whitley 1951) which implies that A is greater than B and hence alignment along the axis would be obtained. However one can only reach temperatures of about 0.08°K (Cooke, Whitley and Wolf 1955) and consequently only low degrees of orientation are obtained. If the cerium is diluted with lanthanum it should be possible to reach much lower temperatures but unfortunately in the dilute salt $g_{\parallel}$ is less than $g_{\perp}$ and hence the lowest nuclear state has $I_z = \pm \frac{1}{2}$. Thus again alignment in a plane would be obtained.

The difficulty was overcome by replacing all but approximately 1% of the cerium which included the radioactive isotope ^{141}Ce by neodymium.

In an auxiliary experiment we had shown that concentrated neodymium ethyl sulphate can be cooled to about 0.024°K by adiabatic demagnetization and this salt has already been used to align ^{147}Nd nuclei (Bishop *et al.* 1955 b). The hfs of cerium in this crystal is not expected to be markedly different from that in concentrated cerium ethyl sulphate. Expressions for A and B in the spin Hamiltonian of Abragam and Pryce (1951) may be obtained from the treatment of Elliott and Stevens (1951, 1952). The value of $\langle 1/r^3 \rangle$ where r is the radial coordinate of the $4f$ electron with respect to the nucleus has been given by Bleaney (1955) as $32.5 \text{ \AA}^{-3}$. In order to evaluate A and B explicitly knowledge of the J_z value of the lowest state is necessary. As will be shown later, axial alignment of cerium in this salt is obtained and therefore the state with $J_z = \pm 5/2$ lies lowest. The corresponding values of A and B are

$$A = 0.108 (\mu_N/I) \text{ cm}^{-1},$$

$$B = 0.002 (\mu_N/I) \text{ cm}^{-1}.$$

A pair of these neodymium ethyl sulphate crystals weighing about 2 g was mounted in the low temperature cryostat with their crystalline axes horizontal. These contained about 13 microcuries of ^{141}Ce , prepared by irradiation of stable cerium oxide in the Harwell pile. This arrangement is convenient for the γ -ray distribution measurement since it allows the counters to be mounted in the horizontal plane. For the neodymium ions

$$g_{\parallel} = 3.61, \quad g_{\perp} = 2.05 \quad (\text{Bleaney, Scovil and Trenam 1952}).$$

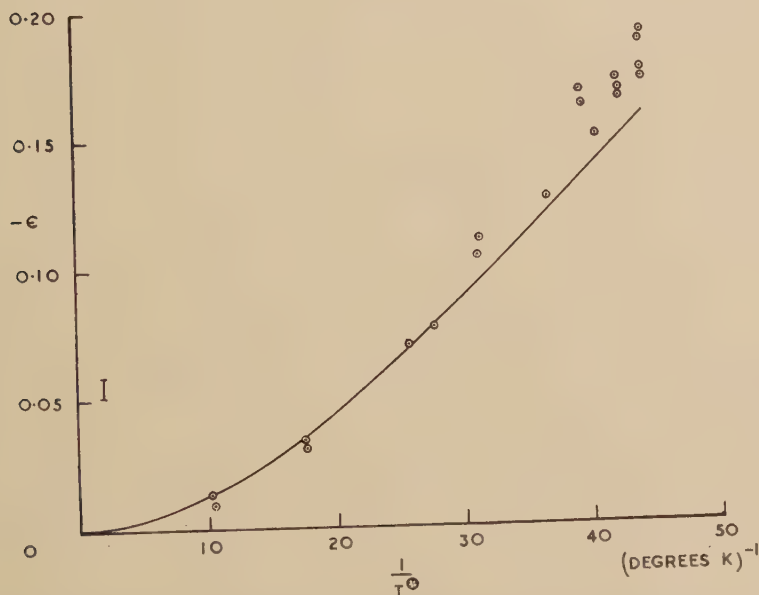
Since these g -factors are of the same order of magnitude the direction of the large magnetic field used in cooling by adiabatic demagnetization is relatively unimportant. The magnetizing field was applied vertically along the $g_{\perp}$ direction. Using values of H/T of 20 kilogauss/ $^{\circ}\text{K}$ magnetic temperatures $T^{\otimes}$ of about 0.024°K were reached. For these crystals it was found that the temperature remained below 0.03°K for about 20 minutes.

§ 5. ANGULAR DISTRIBUTION AND POLARIZATION OF THE γ -RADIATION

On reaching the low temperature the magnet was removed and a pair of NaI (Tl) scintillation counters was mounted round the cryostat. Pulse height selection was used to discriminate against x-radiation and radiation degraded by scattering in the apparatus. The angular distribution was observed at a series of temperatures reached by demagnetizing from different values of the magnetic field. In fig. 2 is shown the dependence on temperature of the γ -ray anisotropy ϵ ; ϵ is defined as $1 - W(0)/W(\pi/2)$ where $W(\theta)$ is the intensity of the γ -radiation emitted at an angle θ with the axis of orientation. It is seen that the anisotropy is negative, whereas Ambler *et al.* found positive values. We therefore conclude that alignment along the crystalline axis is obtained as expected.

Measurements were also made on the plane polarization of the γ -radiation emitted in the plane perpendicular to the alignment axis. These were made with the polarimeter which has been described elsewhere (Bishop and Perez y Jorba 1955) and makes use of the dependence of the differential Compton scattering cross section on the plane of polarization of the γ -radiation. Polarimeter measurements were confined to the lowest temperature region where the effect was largest and these showed the correct general variation with temperature. Defining P' as the ratio of the numbers of quanta scattered in the directions perpendicular to ($N_{\perp}$)

Fig. 2



The dependence of γ -ray anisotropy (ϵ) from aligned Cerium 141 nuclei on $1/T$ ($^{\circ}\text{K}$).

and parallel to ($N_{\parallel}$) the axis of alignment, we find that for $1/T$ of 40 P' is 1.58. This ratio P' is connected with the polarization of the radiation P by the relation

$$P = (1 - P')(R + 1)/(1 + P')(R - 1).$$

P is defined as $(I_{\perp} - I_{\parallel})/(I_{\perp} + I_{\parallel})$ where $I_{\perp}$ and $I_{\parallel}$ are the intensities of the radiations polarized with their electric vectors perpendicular to and parallel to the axis of alignment, and R is the analysing ratio for the polarimeter and depends on its geometry and the γ -ray energy. In this case R is believed to be about 6. This value has been estimated from the known behaviour of the polarimeter. Experiments to test the validity of this assumption are at present in progress.

§ 6. INTERPRETATION OF RESULTS

The intensity of radiation $W(\theta, \phi)$ emitted at an angle θ to the axis of orientation from a system of nuclei can be derived from the expressions given by Biedenharn and Rose (1953) for polarization-direction correlation between successive γ -rays. ϕ is the inclination of the electric vector to the plane containing the axis of orientation and the direction of propagation. The parameter $F_\nu(L_{1j1j})$ which describes the first γ -ray is replaced by the orientation parameter $B_\nu U_\nu$; B_ν describes the orientation of the initial state (in this case the ground state of ^{141}Ce) and U_ν depends upon the spins involved in the unobserved transitions preceding the γ -ray (in this case the β -transition) (Gray and Satchler 1955). For low degrees of orientation where B_2 is the only significant term and for dipole transitions with admixtures of quadrupole radiation this expression reduces to

$$W(\theta, \phi) = 1 + B_2 U_2 F_2(1, I_0, I_1) P_2(\cos \theta) (1 + 3\alpha\delta + 3\beta\delta^2) \\ + \frac{1}{2} B_2 U_2 F_2(1, I_0, I_1) P_2^2(\cos \theta) \cos 2\phi (1 - \alpha\delta + 3\beta\delta^2).$$

$P_2(\cos \theta)$, $P_2^2(\cos \theta)$ are Legendre and associated Legendre polynomials. δ is the ratio of the amplitudes of quadrupole to dipole radiation. It should be noted that ϕ as defined above is different from that used by Biedenharn and Rose, being complementary to their angle.

$$\alpha = 2(-)^{I_1 - I_0 - 1} \sqrt{15(2I_1 + 1)} G_2(1, I_0, I_1) / 3F_2(1, I_0, I_1) \\ \beta = F_2(2, I_0, I_1) / 3F_2(1, I_0, I_1).$$

F_2 and G_2 are functions tabulated by Biedenharn and Rose (1953). σ takes the values $+1$, -1 for magnetic and electric dipole transitions respectively.

Using this expression one can first of all deduce from the signs of P and ϵ that the transition is principally magnetic (either M1 with a small admixture of E2 or M2 with a small admixture of E1). The second choice is unlikely both because the internal conversion coefficient and the lifetime of the state favours an M1 assignment and also because the value of δ required leads to rather small predicted values for the anisotropy at any temperature.

From the value of $+1.58$ for P' at $1/T^\otimes = 40$ we find that P is -0.30 . Combining this with the value of -0.157 for ϵ at the same temperature we find that δ is $+0.08 \pm 0.02$ for the E2/M1 mixing ratio. This result depends only on the spin assignments $7/2$ and $5/2$ for the initial and final states of the γ -ray transition and is independent of any possible perturbation of the states. The experimental error arises from the statistical errors in P' and ϵ and in uncertainty in the value of R . The theoretical curve shown in fig. 2 has been calculated using this value of δ and has been adjusted to pass through the point $\epsilon = -0.066$ at $1/T^\otimes = 25$. This point was chosen because for lower temperatures the difference between $T^\otimes$ and T although unknown may be considerable and for higher temperatures the anisotropy becomes too small to be measured accurately. The deviation of the points from the curve in the low temperature region is attributed

to differences between $T^\otimes$ and T . With this value for δ and a value of 0.066 for $1/T^\otimes = 25$ we find that $B_2 U_2 F_2 (1, I_0, I_1)$ is 0.081 ± 0.017 . In this way we find that B_2 is 0.302 ± 0.066 if the β -transition involves a spin change $i_\beta = 1$ and 0.246 ± 0.054 if the spin change $i_\beta = 0$. From the corresponding values of $\beta (= A/2kT)$ we find that A is $0.0231 \pm 0.0059 \text{ cm}^{-1}$ ($i_\beta = 1$) or $0.0201 \pm 0.0049 \text{ cm}^{-1}$ ($i_\beta = 0$). From the calculated values for AI/μ (see § 4) we find that the nuclear moment of ^{141}Ce is 0.75 ± 0.20 ($i_\beta = 1$) or 0.66 ± 0.16 ($i_\beta = 0$) nuclear magnetons. The large errors here arise principally from uncertainty in the value of R and include an allowance of 10% for the possible difference between absolute and magnetic temperatures.

These figures for the nuclear moment are markedly higher than that obtained by Ambler *et al.* Part of this difference can be attributed to the effect of E2/M1 admixture whose neglect would lead these authors to conclude a smaller value for this moment. If one takes our figure for δ and recalculates their value for the nuclear moment a figure of 0.27 nuclear magnetons is obtained, which is still considerably smaller than our value.

The remaining difference can arise from two possible effects. Either the calculation for the hfs in cerium magnesium nitrate is in error due to interaction between the cerium ions which has not been taken into account or the 145 kev state in ^{141}Pr experiences a perturbation during its lifetime due to interaction with extra-nuclear fields. It seems plausible that such a perturbation could have a greater effect on nuclei oriented in a plane, as in the double nitrate salt where the lower hfs levels are separated by about 0.002 cm^{-1} , than on nuclei oriented along an axis as in the ethyl sulphate where the separation is 0.01 cm^{-1} . In this case we can estimate the magnitude of the perturbation if it is assumed to be negligible for the ethyl sulphate. If we define an attenuation coefficient λ by

$$W(\theta) = 1 + \lambda B_2 U_2 F_2 (1 + 3\alpha\delta) P_2(\cos \theta)$$

where λ takes the value 1 for no perturbation and 0 for complete destruction of the orientation, then it is found for the double nitrate that λ is about 0.5. In view of the uncertainty as to whether a perturbation is also present in cerium ethyl sulphate we must conclude that our values for the nuclear moment represent lower limits.

§ 7. CONCLUSIONS

It is concluded from these experiments on the orientation of ^{141}Ce in a crystal of cerium ethyl sulphate that the 145 kev γ -ray transition excited in its decay is predominantly M1 with a small E2 admixture as is suggested by the Coulomb excitation experiments (see § 2). The value of the mixing ratio δ is 0.08 ± 0.02 . The nuclear moment of ^{141}Ce is 0.75 ± 0.20 or 0.66 ± 0.16 nuclear magnetons depending on whether the β -transition involves a spin change of 1 or 0 and assuming that there is no perturbation of the 145 kev state. There is a difference between the nuclear moment

obtained in these measurements and in those of Ambler *et al.* Part of this can be accounted for by the presence of the E2/M1 mixture and the remainder must be attributed either to a perturbation of the nuclear state in the cerium magnesium nitrate, or to the existence of some other form of coupling not taken account of in the calculation of B in the double nitrate crystal, possibly due to the presence of neighbouring cerium ions.

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CXLI. *The Lattice Spacings of Solid Solutions of Titanium, Vanadium, Chromium, Manganese, Cobalt and Nickel in α -Iron*

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SUMMARY

Accurate measurements have been made of the lattice spacings of dilute solid solutions of titanium, vanadium, chromium, manganese, cobalt and nickel in α -iron. In all cases the formation of the solid solution results in an expansion of the iron lattice, and for equal atomic percentages of the solutes, the expansions are in the order $\text{Ti} > \text{V} > \text{Cr}$ and $\text{Co} < \text{Ni} < \text{Cu}$. Manganese behaves anomalously, its effect being nearly the same as that of chromium. With titanium and vanadium as solutes there are negative deviations from Vegard's law, whilst for the remaining solutes the deviations are positive. These results, together with previous work on nickel alloys, are discussed, and some of the effects are ascribed to exchange repulsion between nearly filled d shells.

§ 1. INTRODUCTION

THE object of the present work was to measure the lattice spacings of the solid solutions in α -iron formed by the elements from titanium to nickel, and to compare the results with those obtained by Hellowell and Hume-Rothery (1954) for the solid solutions in ruthenium formed by the corresponding elements in the Second Long Period, and with those obtained by different workers for solid solutions in nickel.

§ 2. EXPERIMENTAL

(a) *Metals Used*

The iron used was special high purity H iron prepared at the National Physical Laboratory, and obtained from the Pure Metals Committee of the Department of Scientific and Industrial Research; we have to thank Dr. H. Sutton for his help in this connection. The analysis indicated a purity of the order 99.969%, the chief impurities being (in weight percentages) carbon 0.006, manganese <0.005, nickel 0.006, copper 0.004, and aluminium 0.004. The titanium used was I.C.I. Grade A iodide metal whose purity was of the order 99.7%, the principal impurity being iron and therefore harmless. The vanadium used was supplied by Messrs. Johnson Matthey & Co. Ltd., and was in the form of beads of purity 99.7–99.8%. For the work on *iron-chromium*, *iron-manganese* and *iron-nickel* alloys,

* Communicated by the Authors.

use was made of bars of specially pure alloys prepared for work at the National Physical Laboratory, and we must thank Dr. N. P. Allen for presenting these, some of which were used as provided, whilst others were remelted with the high-purity iron. For the *cobalt alloys*, use was made of a master alloy prepared by arc-melting by Messrs. Johnson Matthey & Co. Ltd., and the purity was of the order 99.93%, the chief impurities being nitrogen, oxygen and nickel.

(b) Preparation of Alloys

The iron-titanium alloys were prepared in an argon arc-melting furnace, the earlier melts being carried out at the R.A.E. Farnborough, by the kindness of Mr. L. G. Carpenter. Later alloys were prepared by arc-melting in our own laboratory. Some of the iron-vanadium alloys were prepared by arc-melting at the R.A.E., whilst all other alloys were prepared by melting in an H.F. induction furnace under low pressure of hydrogen, with full precautions of evacuation and degassing. The crucibles used were of recrystallized alumina or thoria, and the alloys were allowed to solidify in the furnace. For the cobalt alloys the hydrogen was pumped off after melting, and solidification took place *in vacuo*.

Specimens from the ingots were homogenized by heating for 3-5 days at 1200°-1250°C in alumina collars contained in silica tubes which were either evacuated or filled with argon to an extent producing a pressure of about 1 atmosphere at the annealing temperature. After homogenization the alloys were air-cooled, except that the iron-nickel and iron-manganese alloys were quenched in order to prevent decomposition on cooling. The N.P.L. alloys had already been homogenized, but were re-annealed and quenched, except for the iron-chromium alloys which were used as received.

(c) X-Ray Methods

For the alloys with titanium, vanadium, chromium and cobalt, filings were prepared and strain annealed at 600°-800°C, whilst for the iron-manganese and iron-nickel alloys the treatments were at 460°-700°C in order to avoid the formation of 2-phase ($\alpha + \gamma$) structures. The filings of alloys with titanium, vanadium and manganese reacted with silica and were therefore placed in alumina protectors inside silica tubes for the strain annealing treatment, and precautions taken to remove adsorbed gas before sealing off. Owing to the ferromagnetic nature of the alloys possible fragments of the file could not be removed magnetically, but as the alloys were soft it is improbable that appreciable errors were introduced in this way, and the work of Owen and Liu (1949) has shown that filings and solid specimens of iron-nickel alloys give identical lattice spacings.

The x-ray specimens were prepared in the form of thin cylinders, using Canada Balsam as adhesive. The powder photographs were taken in two 19 cm Unicam cameras which were checked against one another, the

exact lattice spacings being determined by standard extrapolation methods. The coefficient of expansion of iron was used to correct the values to refer to 20°C and all results are given in kx units, uncorrected for refraction, and subject to this condition should be accurate to better than ± 0.0001 kx so far as x-ray technique is concerned. The standard wavelengths on which the present results are based are :

Cobalt $K\alpha_1$ $\lambda = 1.78529$ kx,

$K\alpha_2$ $\lambda = 1.78919$ kx.

Table 1. Results for Iron Alloys at 20°C

Solute element	Atomic % solute	Lattice parameter kx	Notes
Pure Fe		2.8604(3)	Mean of seven determinations
Ti	0.64	2.8609(5)	} Arc-melted alloys
	0.76	2.8611(5)	
	0.85	2.8615(1)	
	1.27	2.8607(1)	
	1.40	2.8625(6)	
	1.62	2.8633(6)	
	1.64	2.8629(2)	
	2.26	2.8652(8)	
V	2.87	2.8632(8)	} Arc-melted alloys
	3.85	2.8643(3)	
Cr	1.31	2.8610(0)	} N.P.L. alloys
	2.26	2.8616(0)	
	3.41	2.8621(8)	
	5.37	2.8632(8)	
Mn	0.81	2.8604(5)	Induction-melted alloy
	1.08	2.8607(6)	N.P.L. alloy
	1.09	2.8606(1)	} Induction-melted alloys
	1.79	2.8610(6)	
	1.95	2.8613(2)	N.P.L. alloy
	2.35	2.8615(4)	Induction-melted alloy
	3.09	2.8618(1)	N.P.L. alloy
Co	2.19	2.8606(8)	} Induction-melted alloys
	4.14	2.8608(0)	
	6.22	2.8608(5)	
	9.16	2.8610(9)	Master alloy
Ni	1.91	2.8614(8)	} N.P.L. alloys
	2.66	2.8619(0)	
	3.87	2.8624(6)	
	4.96	2.8627(2)	

§ 3. THE LATTICE SPACING OF PURE IRON

The lattice spacing of the pure H iron was determined as $2.8604(3)$ kx at 20°C in good agreement with values of between 2.8601 kx and 2.8605 kx obtained by different previous workers after reduction to 20°C .

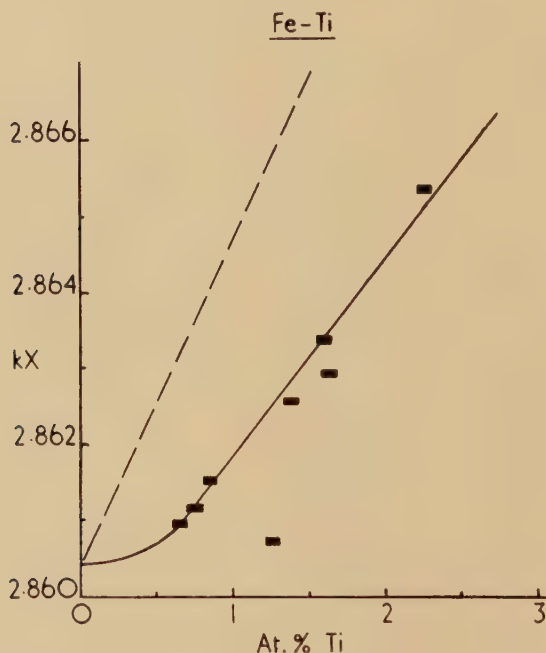
§ 4. LATTICE SPACINGS OF SOLID SOLUTIONS IN IRON

The lattice spacings obtained in the present work are summarized in table 1 and the following points may be noted.

(a) *Iron-Titanium*

No previous accurate data existed for this system, and the present results are shown in fig. 1. Of the eight points, seven lie on a straight line, and one is incorrect for reasons which are not understood. The

Fig. 1



straight line does not, however, extrapolate to the value for pure iron, and the first additions of titanium produce a relatively small lattice expansion. This effect is not understood, and is too great to be due to intercrystalline adsorption of solute. It is also too great to be ascribed to oxidation or nitriding during melting which might convert titanium into oxide or nitride remaining in a form which was dissolved during chemical analysis and so give an apparent titanium content higher than that of the solid solution. The effect resembles that found by Axon and Hume-Rothery (1948) for solid solutions of high purity (99.97%) magnesium in aluminium, and here later work by Poole and Axon (1951)

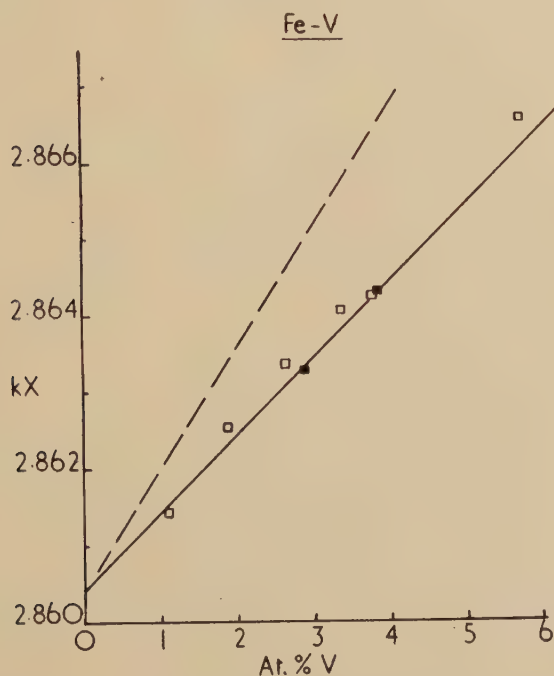
showed that the effect was removed by using more pure (99.98%) magnesium. It is probable, therefore, that the effect is due to traces of impurity in the alloys, but in spite of much work it is not understood.

In fig. 1 the dotted line shows the results expected if Vegard's law were obeyed, and the lattice spacing of titanium were that of the β (body-centred cubic) modification corrected to refer to room temperature. In the iron alloys there is a contraction compared with Vegard's relation, and there would still be a slight contraction if the straight line part of the present curve were displaced to pass through the value for pure iron.

(b) Iron-Vanadium

In fig. 2, the full points show the lattice spacings of the two arc-melted alloys, and these lie on a straight line passing through the value for pure iron. The open points refer to alloys which were accidentally very

Fig. 2



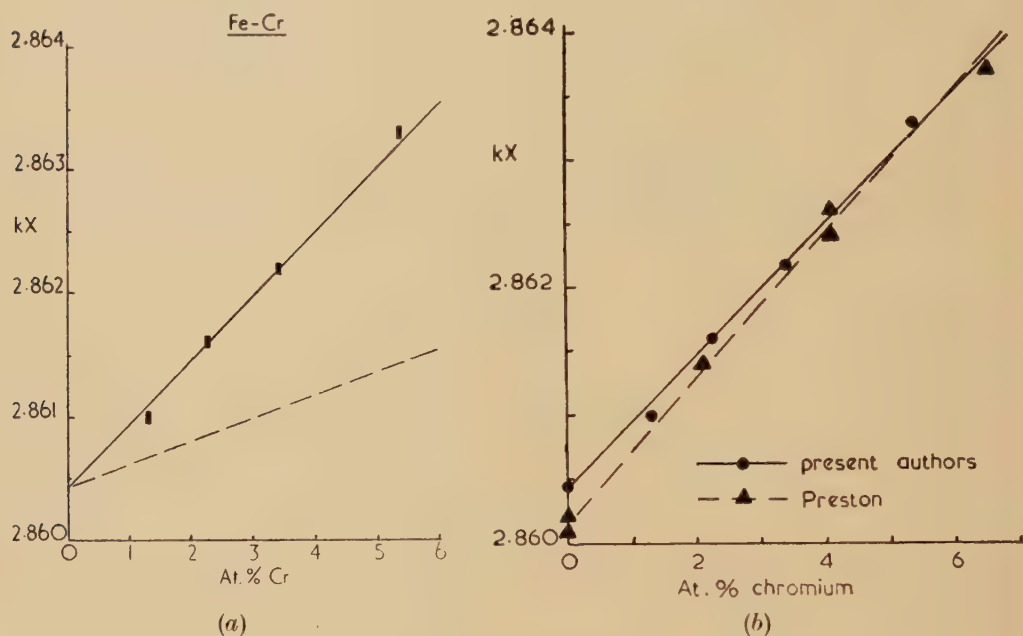
slightly contaminated by tantalum and which are therefore not included in table 1. The dotted line shows the lattice spacings expected according to Vegard's law, and there is again a marked contraction.

(c) Iron-Chromium

Figure 3(a) shows the results obtained for iron-chromium alloys, whilst the dotted line shows the lattice spacings expected according to Vegard's Law. Figure 3(b) shows the present results compared with those found by Preston (1932). When allowance is made for the slightly

different values for pure iron, there is good agreement between the two determinations although the present results indicate a slightly smaller increase in lattice spacing. In this system the lattice spacings are greater than would be expected from Vegard's law.

Fig. 3



(d) Iron-Manganese

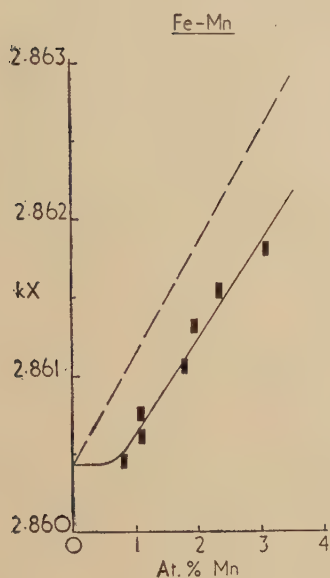
Figure 4 (a) shows the results obtained for iron-manganese alloys, together with those of Troiano and McGuire (1943) (fig. 4 (b)). The points for the seven alloys examined in the present work lie on a straight line to a very high degree of accuracy, but this straight line extrapolates to a point about 0.0004 kX below the value for pure iron, although the effect is not so pronounced as for the iron-titanium alloys. If allowance is made for the slight difference in the lattice spacings of the iron used, the present results agree well with those of Troiano and McGuire which also show an abnormality at low concentrations. The data suggest a slight contraction compared with those expected from Vegard's law, shown by the dotted line in fig. 4 (a) although this difference would almost disappear if the straight line portion of the curve were displaced parallel to itself so as to pass through the value for pure iron.

(e) Iron-Cobalt

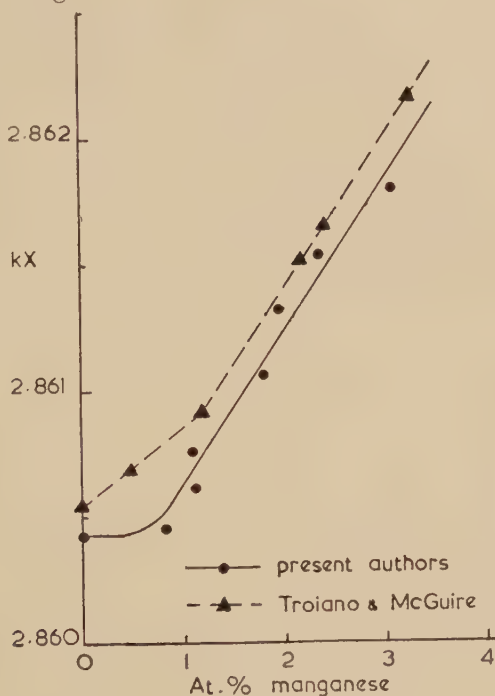
Figure 5 (a) shows the results obtained for iron-cobalt alloys together with those of Rodgers and Maddocks (1939), and of Ellis and Greiner (1941) (fig. 5 (b)). The lattice distortion is very small, but is greater

than that found by Ellis and Greiner, although in good agreement with that of Rodgers and Maddocks if allowance is made for the difference between the lattice spacings of the iron used. In fig. 5 (a) the upper

Fig. 4

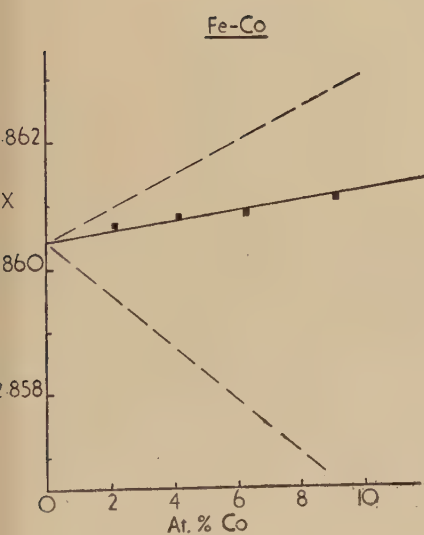


(a)

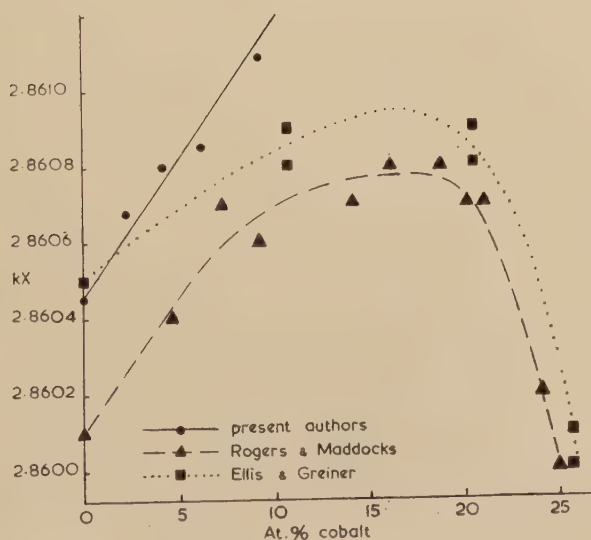


(b)

Fig. 5



(a)



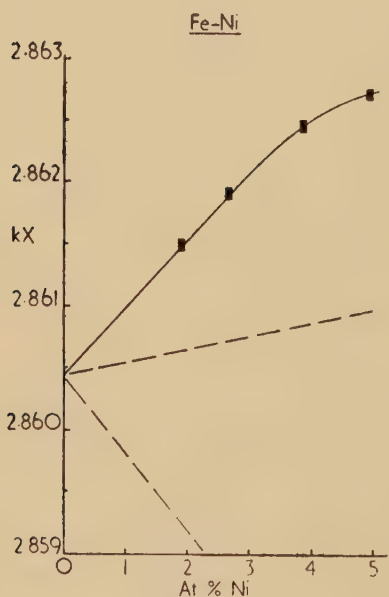
(b)

dotted line shows the results expected if Vegard's law held, and the atomic diameter of cobalt were that observed in α - and β -cobalt. This, however, refers to structures with coordination no. 12, and should therefore be reduced by 2.5%* in order to give the values appropriate to a hypothetical body-centred cubic modification of cobalt. If this correction is made, the results show an expansion compared with Vegard's relation (lower dotted line, fig. 5 (a)), although there is a slight contraction if no correction is made.

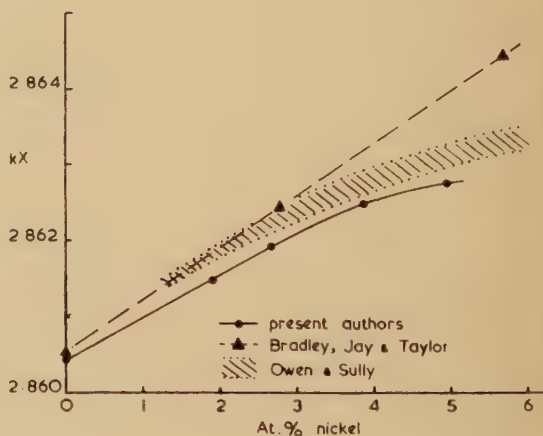
(f) *Iron-Nickel*

Figure 6 (a) shows the results obtained for iron-nickel alloys, together with those of Bradley, Jay and Taylor (1937), and of Owen and Sully (1941) (fig. 6 (b)). When allowance is made for the different values for pure iron, the present results show a smaller expansion of the lattice

Fig. 6



(a)



(b)

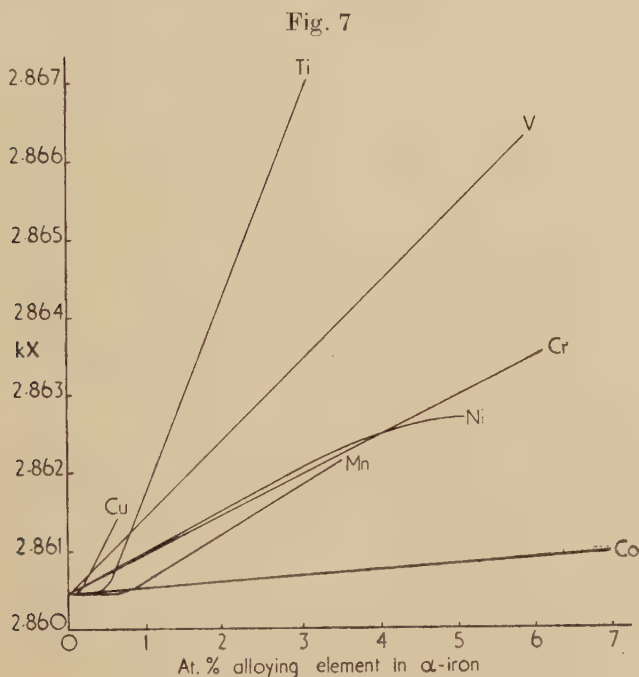
than was claimed by Bradley and Jay, but are in good agreement with the data of Owen and Sully. The lattice spacing/composition curve is at first a straight line, and then shows a decrease in the rate of expansion. The initial straight portion of the curve shows an appreciably greater rate of expansion than would be expected from Vegard's law for a hypothetical body-centred cubic nickel with an interatomic distance equal to that of face-centred cubic nickel (upper dotted line, fig. 6 (a)), whilst if the interatomic distance in nickel is corrected to allow for the

* See p. 1304 for details.

contraction on change of coordination no. $12 \rightarrow 8$, the positive deviation from Vegard's law is even greater (lower dotted line, fig. 6 (a)).

§ 5. DISCUSSION

Figure 7 shows the lattice spacing/composition curves obtained in the present work, together with that of Norton (*A.S.M. Handbook*) for the restricted solid solution of copper in α -iron. Considered empirically, the



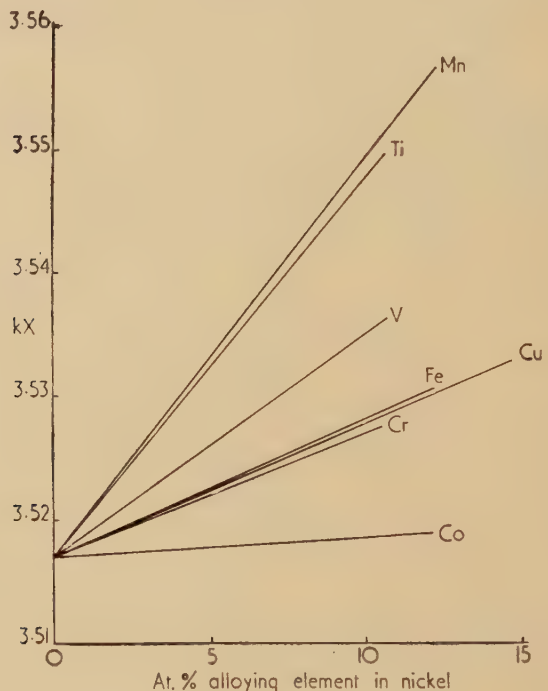
curves fall into two sequences, in which at a given atomic percentage of solute, the lattice expansions are in the order $Ti > V > Cr$ and $Co < Ni < Cu$. The behaviour of Mn is anomalous, its effect being nearly the same as that of chromium.*

Figure 8 shows the lattice spacing/composition curves of the same elements dissolved in nickel, and here the effects are in the same general order $Ti > V > Cr$ and $Co < Cu$, although the lattice of nickel is very slightly expanded by cobalt, in contrast to the order $Co < Ni < Cu$ found in the iron alloys. For the nickel alloys, the abnormal behaviour of manganese is more pronounced, and the Ni-Mn curve lies close to that for Ni-Ti.

* Detailed discussion of the curve for manganese is difficult because of the abnormality at low concentrations. If the Fe-Mn curve were shifted parallel to itself so as to pass through the value for pure iron it would lie between the curves for Fe-V and Fe-Cr.

It is also instructive to compare the data for the iron alloys with those found by Hellowell and Hume-Rothery (1954) for solid solutions of the corresponding elements of the Second Long Period in the close-packed hexagonal solvent ruthenium. Here at a given atomic composition,

Fig. 8



the expansions of the c lattice spacing of ruthenium are in the order $\text{Zr} > \text{Nb} > \text{Mo}$, and $\text{Rh} < \text{Pd}$, the effect of Rh being small,* and that of Pd slightly less than that of Mo. Quite different effects are shown by the a spacings, which are contracted by Zr, and very slightly contracted by Rh, whilst Nb and Mo produce large and roughly equal expansions, and Pd a less marked but considerable expansion. Considered empirically, therefore, it is the effects for the c spacing of ruthenium which resemble those found in the present work for solid solutions in iron.†

To discuss the curves for iron and nickel in greater detail, correction must be made for the effects of coordination numbers. The data for iron (Basinski, Hume-Rothery and Sutton 1955), and manganese (Basinski and Christian 1954) suggest that Goldschmidt's empirical

* Data for Te alloys are lacking.

† In the ruthenium alloys the effects of the solutes upon the volume of the unit cell are such that Zr causes a contraction, Rh has no effect, whilst the remaining elements cause expansions in the order Nb, Mo (approx. equal) $>$ Pd.

factor of 3% contraction in atomic diameter on passing from face-centred cubic to body-centred cubic coordination is slightly too great, and that a contraction of 2.5% is in better agreement with the facts. Using this correction, the values in table 2 are obtained for the atomic diameters of the elements concerned.

Table 2

Element	Ti	V	Cr	Mn	Fe	Co	Ni	Cu
Atomic diameter in face-centred cubic	2.89* to 2.94	2.70‡	2.56‡	2.61	2.54	2.50	2.49	2.55
Atomic diameter in body-centred cubic	2.84‡	2.62	2.49	2.54	2.48	2.44‡	2.43‡	2.49‡

* Values for the two close distances of approach in the close-packed hexagonal structure of α -Ti.

† Value from extrapolation of lattice spacings of solid solutions in β -Ti.

‡ Values calculated by assuming 2.5% contraction on passing from face-centred cubic to body-centred cubic.

Using these values, the data for Fe-Ti and Fe-V show a contraction compared with Vegard's law, whilst those for Fe-Cr, Fe-Co, Fe-Ni and Fe-Cu show an expansion. This difference is reflected in the equilibrium diagrams of the systems concerned. In the systems Fe-Ti and Fe-V there are intermetallic compounds or intermediate phases of high melting point, which suggest strong bonding of the two kinds of atom. In the system Fe-Cr, the equiatomic σ phase is stable only below 815°C, whilst in the remaining systems there are no intermediate phases apart from superlattices stable only at low temperatures.

Although there are departures from Vegard's law, the lattice expansions $\text{Ti} > \text{V} > \text{Cr}$ are in the same order as that expected from the atomic diameters, but from this stage onwards the order is quite different, and the behaviour of manganese is clearly anomalous. For the elements succeeding iron, the lattice expansions are in a well defined sequence $\text{Co} < \text{Ni} < \text{Cu}$, and bear no relation to the atomic diameters which are all nearly the same, and to a higher degree of accuracy are in the order

$$\text{Ni } 2.43 < \text{Co } 2.44 < \text{Fe } 2.48 < \text{Cu } 2.49.$$

We may use the term 'outer electrons' to denote all the electrons outside the argon shell, and 'electron number' to describe the number of electrons per atom concerned in metallic bonding. § In the 'collective

§ This has usually been called the valency of the atom in the metal concerned, but the term has been used with so many meanings that we prefer to speak of the electron number.

electron theories many authors now regard all of the outer electrons as taking part in the bonding process, so that the electron number is equal to the number of outer electrons. From this viewpoint, the break in the sequence of physical properties in the middle of the transition elements is the result of an increase in the repulsive part of the wave functions of the d-electrons, which becomes very marked on passing through Group VIII (Co→Ni, Rh→Pd, Ir→Pt). In the *resonating bond theories*, the cohesion in crystals of the transition metals results from the resonance of hybrid spd bonds, and the breaks in the physical property sequences are the result of some electrons entering atomic or non-bonding orbitals. In the Pauling (1938) hypothesis the electron number is regarded as increasing from 1 to 5 on passing from K→V, and then as remaining constant at 5.78 on passing from Cr→Ni. As pointed out by Hume-Rothery, Irving and Williams (1951) this scheme is not in agreement with the physical property sequences, and these authors suggest that, in the First Long Period,* the electron numbers increase from 1 to 4 on passing from K→Ti, with an uncertain value for vanadium in Group VA, and a value for chromium which is lower than those for molybdenum and tungsten in the later Periods. The abnormal properties of manganese were regarded as associated with more than one electronic state, whilst the relatively large interatomic distance in γ -Mn and the low melting point of δ -Mn were associated with an electron number 2, corresponding with the very stable divalent Mn^{2+} compounds. Later work by Basinski and Christian (1954) has shown that δ -Mn has also a relatively large closest interatomic distance. All the physical properties suggest a rise in electron number on passing from Mn→Fe, together with a very gradual decrease in the sequence Fe→Co→Ni. Hume-Rothery, Irving and Williams pointed out that the electron numbers suggested by the physical properties ran parallel to the predominant valencies in the chemistry of these elements.

In an early discussion Mott (1944) regarded copper, silver and gold as consisting of 'hard ions' pulled into contact by the s valency electrons, with a simultaneous additional cohesion of the van der Waals type arising from the perturbation of the outer d electrons of the ion, and this view has been discussed by one of us (Hume-Rothery 1955 a). Recently Mott (1953) has suggested that this concept can be applied to metals at the end of the transition series, the van der Waals perturbation being treated as an excitation of the d→p-type. This picture, although not its mathematical details, is not so very different from that of the resonating bond theories, since each imagines a core of repulsive electrons, and each involves s, p and d states for the cohesion. On passing back from Ni→Co→Fe the d electron clouds become larger and more loosely

* In the Second and Third Long Periods the electron numbers increase from 1 to 6 on passing from Groups IA-VIA, remain high in Groups VII and VIIIA, and then fall rapidly on passing to Groups VIIIB and VIIIC.

held, and the van der Waals perturbation forces increase until they become so large that they break into normal metallic cohesion.

When we try to combine the different concepts, we may regard the almost exactly equal atomic diameters in the elements Fe, Co, Ni, Cu as the result of the following opposing tendencies:

(1) With increasing atomic number the 3d electrons become more firmly held, and the electron clouds of both 3d and 4s electrons contract, and thus tend to produce a smaller atomic diameter.

(2) The contraction and firmer binding of the 3d electrons means that they are less easily perturbed, and this reduces the attractive force, whether the latter is regarded as resulting from spd hybridization, or from van der Waals perturbation of the d electrons. At the same time the filling up of the (3d) shell increases the exchange repulsion for a given degree of overlap of the electron clouds, and these two effects tend to increase the atomic diameter.

(3) The fall in electron number suggested by the chemical valencies on passing from Fe→Cu weakens the attraction. In so far as this fall results from the withdrawal of attractive d electrons it is perhaps really included in (1) and (2) above.

Using the low electron numbers of H-R, I and W, a considerable proportion of the d electrons round each atom in an iron crystal is repulsive or non-bonding, but the number of these repulsive electrons (5 to 6 per atom) is only slightly more than one-half of that required to fill the (3d)¹⁰ group, and so considerable overlap of the cores can take place. The atomic diameter of copper when corrected for coordination number is almost equal to that of iron (table 2) so that if we regard copper as consisting of hard ions with completed (3d)¹⁰ groups in contact, the insertion of a copper atom into the iron lattice will produce an overlap of the completely filled (3d)¹⁰ electron cloud with the (3d) electron cloud of the iron atoms, and the resulting exchange repulsion may be regarded as the cause of the marked expansion of the iron lattice, this effect being reinforced by a weakening of the van der Waals attraction,* and a decrease in the electron number of the solute (Cu) as compared with the solvent atom (Fe). With nickel as solute we may expect the same effects but to a lesser degree. The (3d) core of a nickel atom in solution in iron is probably nearly although not completely† filled, and although the unfilled states permit bonding, any marked overlapping will result in exchange repulsion. With cobalt as solute the (3d) core is less completely filled, and exchange repulsion will be less marked. The curves in fig. 7 show clearly that although the expansions produced by a given atomic percentage of the solutes are in the order Co<Ni<Cu,

* Or alternatively a decrease in the perturbation which enables metallic bonding to occur.

† It is possible that the holes in the d shell of the nickel atoms are filled by electrons from the iron atoms (see p. 1308).

the effect of cobalt is very small, and we may conclude that the exchange repulsion increases enormously when nickel is reached. The fact that cobalt produces a slight lattice expansion of iron, corresponding to a marked positive deviation from Vegard's law, can readily be understood as partly due to the exchange repulsion effect, and partly due to the factors outlined in (2) above. The atomic diameter of iron (2.48 kx) results from an electron number slightly greater than that of cobalt, reinforced by the van der Waals attraction of the iron d shell core, which is larger and more easily perturbed than the d shell core of cobalt. The substitution of cobalt for iron thus weakens the van der Waals attraction and increases the exchange repulsion.

If the above interpretation is correct, we can understand why the lattice expansions of iron produced by Co, Ni and Cu bear no relation to the atomic diameters of the solutes. We can also perhaps understand why the lattice of nickel is slightly expanded by cobalt, and more so by iron, because in nickel the (3d) shell is so nearly filled that exchange repulsion may predominate over increased attraction when the larger (3d) electron clouds of cobalt or iron are substituted.* The same interpretation enables us to understand why the lattice of nickel is expanded by copper more than would be expected from Vegard's law, because, although the 3d electron cloud of a copper atom is smaller than that of a nickel atom, the fact that the (3d)¹⁰ sub-group is filled in copper means that intense repulsion will result from a relatively small degree of overlap, particularly since the magnetic evidence shows that the (4s) electrons of the copper atoms fill the holes in the (3d) shells of the nickel atoms and hence make the latter more liable to give rise to exchange repulsion. When nickel is dissolved in copper, there is a positive deviation from Vegard's law which can be understood as the result of exchange repulsion, but the lattice is contracted and not expanded. It is perhaps significant that, as emphasized by Coles (1952), nickel when dissolved in copper gives paramagnetic alloys, indicating that the (3d) shells of the nickel atoms are not being filled by electrons from the copper atoms.

From the above viewpoint, the distortion of the iron lattice produced by manganese is anomalous, and the effects of cobalt, nickel and copper result from the interplay of several factors. The effects of titanium, vanadium and chromium are more simple, and of these three elements the size factor is really favourable only for vanadium and chromium. In recent work in this laboratory by A. Hellawell, the liquidus and solidus curves of this series of iron-rich alloys have been

* The magnetic evidence shows that the saturation moment of nickel is increased by solution of cobalt or iron, but it is not possible to say whether this is accompanied by a filling of the holes in the 3d band of the nickel atoms. In view of the strong tendency to fill the d shell in the last element of each transition series, it is possible that the substitution of an earlier element (Co, Fe, Mn . . .) always tends to fill the 3d shell of nickel, and so to increase the exchange repulsion. In this case the increase in saturation moment would result from the solute atoms.

determined accurately, and the results suggest that in dilute solutions a given atomic percentage of vanadium depresses the melting point by slightly less than twice* the depression caused by the same atomic percentage of chromium. The present work shows that the lattice expansion for a given atomic percentage of vanadium is slightly less than twice that for the same atomic percentage of chromium. For these two systems, therefore, the lattice expansions and depressions of melting point are related by the same factors, as is the case for some silver alloys examined by one of us (Hume-Rothery 1955 b). This correspondence, however, no longer holds for the system iron-titanium where the lattice distortion is much greater.

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* The experimental data could be reconciled with a factor of exactly 2.

*CXLII. On the Application of Eigenfunction Expansions to the Problem
of the Thermal Instability of a Fluid Sphere Heated Within*

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SUMMARY

The eigenvalue problem which describes the onset of convection in a fluid sphere heated within is solved by means of expansions of the convective velocity field and temperature in spherical Bessel functions. The Rayleigh number $C_{l,n}$, at which there first appears the n th radial convection pattern with the spherical symmetry of the l th spherical harmonic, can be computed for any l and n by this device, and bracketed to arbitrary accuracy. A table of $C_{l,n}$ for l from 1 through 6 and $n=1$ and 2 is compared with Chandrasekhar's values of $C_{l,1}$ obtained by a variational method. A similar attack is shown to succeed on the classical Rayleigh-Benard problem of convection in a horizontal layer of fluid. Finally, the class of problems which should be amenable to such an attack is delineated.

§ 1. INTRODUCTION

THE linear partial differential equations which describe the onset of convection in a fluid sphere heated within were first derived by Wasiutinsky (1946). These equations constitute an eigenvalue problem, the eigenvalues being the Rayleigh numbers at which various patterns of convection can set in. Jeffreys and Bland (1951) computed the lowest eigenvalues by a variational procedure, and by means of a more incisive variational technique Chandrasekhar (1952; this paper will be referred to hereafter as I) was able to obtain quite accurate estimates of the lower eigenvalues. Variational estimates of eigenvalues suffer from two disadvantages: they are upper bounds, which in principle cannot bracket the true values; and the higher eigenvalues are inaccessible without inordinate accuracy in the computation of the lower ones. Therefore, in this paper the eigenvalues will be computed by a more laborious technique which does bracket them and which can be used to compute any eigenvalue independently of the others. This method consists of an application of Chandrasekhar's scheme for lowering the order of a high order differential equation. The method exhibits the eigenvalues as the zeros of a certain explicit meromorphic function of a single complex variable and, as a by-product, yields expansions for the

* Communicated by S. Chandrasekhar, F.R.S.

eigenfunctions (the convection currents) in series of spherical Bessel functions. Chandrasekhar's formulation of the problem will be used throughout this paper, and with two exceptions his notation will be followed in detail. Just as his procedure can be extended to spherical shells (Chandrasekhar 1953 b), so can the techniques to be presented here. However, this extension involves laborious computations, so it will be omitted.

§ 2. THE EQUATIONS GOVERNING MARGINAL STABILITY

Chandrasekhar's derivation of the linearized equations for the onset of convection in a fluid sphere heated from within appears in I and will not be reproduced here. In copying his equations, their numbers in I will be given beside their numbers in this paper. In his notation, κ is the thermometric conductivity of the fluid, ν is its kinematic viscosity, $\bar{\rho}$ is its average density, α is its volume coefficient of thermal expansion, and $6\kappa\beta$ is the heat generated per unit volume per unit time in the fluid. The universal constant of gravitation is G , and $\gamma=(4/3)\pi\bar{\rho}\alpha G$, while $\mathbf{L}=-i(\mathbf{r}\times\nabla)$ is the 'angular momentum' differential operator which arises in connection with the spherical harmonics. Finally, u_r is the radial velocity of the fluid, and θ is the deviation of the fluid's temperature from the steady temperature in the absence of convection. Chandrasekhar's equations, based on the principle of exchange of stabilities, are then

$$\kappa\nabla^2\theta=-2\beta ru_r, \quad . \quad . \quad . \quad . \quad (1) \quad (\text{I (24)})$$

and

$$\nabla^4(ru_r)=(\gamma/\nu)\mathbf{L}^2\theta. \quad . \quad . \quad . \quad . \quad (2) \quad (\text{I (34)})$$

Taking R to be the radius of the sphere, the boundary conditions are

$$\theta=0 \text{ at } r=R, \quad . \quad . \quad . \quad . \quad . \quad (3) \quad (\text{I (16)})$$

$$ru_r=0 \text{ at } r=R, \quad . \quad . \quad . \quad . \quad . \quad (4) \quad (\text{I (16)})$$

and $\frac{\partial}{\partial r}(ru_r)=0$ at $r=R$ for a rigid boundary, $. \quad (5) \quad (\text{I (19)})$

or $\frac{\partial}{\partial r}(ru_r)=0$ at $r=R$ for a free boundary. $. \quad (6) \quad (\text{I (23)})$

These equations can be separated by the substitutions

$$ru_r=W(r)Y_l^m(\vartheta, \phi) \quad \text{and} \quad \theta=\Theta(r)Y_l^m(\vartheta, \phi).$$

The resulting differential equations are

$$\frac{1}{r}\left[\frac{d^2}{dr^2}-\frac{l(l+1)}{r^2}\right]r\Theta=-\frac{2\beta W}{\kappa}, \quad . \quad . \quad . \quad . \quad (7)$$

and $\frac{1}{r}\left[\frac{d^2}{dr^2}-\frac{l(l+1)}{r^2}\right]^2rW=\frac{\gamma}{\nu}l(l+1)\Theta. \quad . \quad . \quad . \quad . \quad (8)$

(15) and (16). For example, if the boundary is rigid, this equation is

$$0 = \begin{vmatrix} j_l(\alpha) & \Re[j_l(\alpha e^{i\pi/3}) + (-1)^l j_l(\alpha e^{i2\pi/3})] \\ & \Im[j_l(\alpha e^{i\pi/3}) - (-1)^l j_l(\alpha e^{i\pi/3})] \\ j_l(\alpha) & \Re[e^{i4\pi/3} j_l(\alpha e^{i\pi/3}) + (-1)^l e^{i2\pi/3} j_l(\alpha e^{i2\pi/3})] \\ & \Im[e^{i4\pi/3} j_l(\alpha e^{i\pi/3}) - (-1)^l e^{i2\pi/3} j_l(\alpha e^{i2\pi/3})] \\ j_{l+1}(\alpha) & \Re[e^{i\pi/3} j_{l+1}(\alpha e^{i\pi/3}) + (-1)^l e^{i2\pi/3} j_{l+1}(\alpha e^{i2\pi/3})] \\ & \Im[e^{i\pi/3} j_{l+1}(\alpha e^{i\pi/3}) - (-1)^l e^{i2\pi/3} j_{l+1}(\alpha e^{i2\pi/3})] \end{vmatrix}. \quad (18)$$

Then $C_{l,m} = \alpha^6$. Equation (18) is sufficiently formidable to prompt other approaches.

The Sturm-Liouville theory ensures that F can be expanded as

$$F = \sum_{n=1}^{\infty} A_n j_l(\alpha_{l,n} r), \quad . \quad . \quad . \quad . \quad . \quad . \quad (19)$$

where $\alpha_{l,n}$ is the n th positive root of $j_l(\alpha) = 0$. We shall usually omit the l , writing simply α_n . The boundary condition that F behaves like r^l as $r \rightarrow 0$ ensures that (19) converges uniformly on the interval $[0, 1]$ (Watson 1944, p. 615). Because of this uniform convergence, the first to fourth anti-derivatives of F can be computed by integrating (19) term by term. Therefore eqn. (11) together with the boundary conditions (14) and (15) can be solved exactly for W in terms of F , by virtue of (17). Thus the sixth order problem embodied in (10) and (11), namely

$$\frac{1}{r} \left[\frac{d^2}{dr^2} - \frac{l(l+1)}{r^2} \right]^3 r W = -l(l+1) C_{l,m} W, \quad . \quad . \quad . \quad . \quad (20)$$

can be reduced by four exact integrations to a second order problem. This technique was introduced by Chandrasekhar, who discusses many of its applications (see particularly Chandrasekhar 1954). In I he applies the method to eqn. (20) as outlined above. It should be observed that either F or W can be expanded in a uniformly convergent series of spherical Bessel functions as in (19). We expand F because then the boundary condition remaining after the four exact integrations is the one that takes two possible forms, namely (16), and the necessity for discussing two cases is delayed as long as possible. Also, starting with F facilitates comparison with Chandrasekhar's procedure in I.

The result of solving (11) for W in terms of the F given in (19) is

$$W(r) = \sum_{n=1}^{\infty} \frac{A_n}{\alpha_n^4} j_l(\alpha_n r) + B r^l + C r^{l+2} + \frac{M}{r^{l-1}} + \frac{N}{r^{l+1}}. \quad (21) \quad (\text{I } (71))$$

Note that the A_n in (16) differs from Chandrasekhar's A_n in I by a factor $(2/\pi\alpha_n)^{1/2}$. Boundary condition (13) requires $M = N = 0$, and $W(1) = 0$ implies $B = -C$, so that (21) becomes

$$W(r) = \sum_{n=1}^{\infty} \frac{A_n}{\alpha_n^4} j_l(\alpha_n r) + B(r^l - r^{l+2}). \quad . \quad . \quad (22) \quad (\text{I } (73))$$

The requirement $F(1)=0$ now implies

$$B = -\frac{q}{4} \sum_{n=1}^{\infty} \frac{A_n}{\alpha_n^3} j_{l+1}(\alpha_n), \quad \dots \quad (23) \quad (\text{I } (74))$$

where

$q=2$ for a rigid boundary,

and

$q=-4/(2l+1)$ for a free boundary.

Now (22) can be substituted in (12) and, since (22) converges uniformly if (17) does so, the result can be integrated term by term to produce

$$\frac{F(r)}{l(l+1)C_{l,m}} = \sum_{n=1}^{\infty} \frac{A_n}{\alpha_n^6} j_l(\alpha_n r) + B \left[\frac{r^{l+4}}{4(2l+5)} - \frac{r^{l+2}}{2(2l+3)} \right] + Dr^l + \frac{E}{r^{l+1}}.$$

The boundary condition (15) makes $E=0$, and (14) determines D , so that

$$\frac{F(r)}{l(l+1)C_{l,m}} = \sum_{n=1}^{\infty} \frac{A_n}{\alpha_n^6} j_l(\alpha_n r) + B \left[\frac{r^{l+4}}{4(2l+5)} - \frac{r^{l+2}}{2(2l+3)} + \frac{(2l+7)r^l}{4(2l+3)(2l+5)} \right]. \quad (24)$$

Now (19) and (15) imply

$$\begin{aligned} \sum_{n=1}^{\infty} A_n \left(\frac{1}{l(l+1)C_{l,m}} - \frac{1}{\alpha_n^6} \right) j_l(\alpha_n r) \\ = B \left[\frac{r^{l+4}}{4(2l+5)} - \frac{r^{l+2}}{2(2l+3)} + \frac{(2l+7)r^l}{4(2l+3)(2l+5)} \right]. \end{aligned} \quad (25)$$

Since the Fourier-Bessel expansion of the right side of (25) is easily obtained, (25) determines A_n in terms of α_n , B and $C_{l,m}$.

To obtain A_n in terms of the Fourier-Bessel coefficients of the right side of (25), we use the following standard relations among spherical Bessel functions :

$$\int_0^1 j_l(\alpha_n r) j_l(\alpha_m r) r^2 dr = \frac{1}{2} j_{l+1}^2(\alpha_n) \delta_{mn}, \quad \dots \quad (26)$$

$$\int_0^1 r^{l+2} j_l(\alpha_n r) dr = (1/\alpha_n) j_{l+1}(\alpha_n), \quad \dots \quad (27)$$

$$\int_0^1 r^{l+4} j_l(\alpha_n r) dr = \frac{j_{l+1}(\alpha_n)}{\alpha_n} \left[1 - \frac{2(2l+3)}{\alpha_n^2} \right], \quad \dots \quad (28)$$

and

$$\int_0^1 r^{l+6} j_l(\alpha_n r) dr = \frac{j_{l+1}(\alpha_n)}{\alpha_n} \left[1 - \frac{4(2l+5)}{\alpha_n^2} + \frac{8(2l+3)(2l+5)}{\alpha_n^4} \right]. \quad (29)$$

Multiply (25) by $r^2 j_l(\alpha_n r)$ and integrate from 0 to 1. Using (26) through (29), the result is

$$A_n = B \frac{4\alpha_n(2l+3)l(l+1)C_{l,m}}{j_{l+1}(\alpha_n)[\alpha_n^6 - l(l+1)C_{l,m}]}. \quad \dots \quad (30)$$

If $C_{l,m}$ is known, the corresponding temperature Θ and radial velocity W/r can be computed from (30), (19), (22) and (23). All the results of this

paper follow from (30). In particular if (30) is substituted in (23) and B is cancelled, the following equation for $C_{l,m}$ is obtained :

$$\frac{1}{q(2l+3)} = \sum_{n=1}^{\infty} \frac{l(l+1)C'_{l,m}}{\alpha_n^2[l(l+1)C'_{l,m}-\alpha_n^6]}. \quad (31)$$

§ 4. GENERAL REMARKS ABOUT THE LOCATION OF THE EIGENVALUES

The remainder of this paper, except for §§ 7 and 8, is a discussion of the roots of the equation

$$\psi_l(z) = \frac{1}{q(2l+3)} \quad (32)$$

where

$$\psi_l(z) = \sum_{n=1}^{\infty} \frac{z}{\alpha_{l,n}^2(z-\alpha_{l,n}^6)}. \quad (33)$$

Figure 1 is a graph of $\psi_1(z)$ for real z . If $r_{l,m}$ is the m th positive root of (32), then

$$l(l+1)C'_{l,m} = r_{l,m}. \quad (34)$$

The subscript l will be omitted from $r_{l,m}$ hereafter.

If the interiors of small circles around the simple poles $z=\alpha_n^6$ are deleted, then in any bounded region of the complex plane the series (33) converges uniformly. Therefore (Copson 1935, p. 97) its sum is analytic and can be differentiated term by term. Thence

$$\psi'(z) = - \sum_{n=1}^{\infty} \frac{\alpha_n^4}{(z-\alpha_n^6)^2}. \quad (35)$$

It is clear that if z is real $\psi'(z) < 0$, so (31) has precisely one real root between α_N^6 and α_{N+1}^6 . Furthermore, if $z < 0$, $\psi(z) > 0$ and $\psi(z)$ decreases monotonically from

$$\lim_{z \rightarrow -\infty} \sum_{n=1}^{\infty} \frac{z}{\alpha_n^2(z-\alpha_n^6)}.$$

Since

$$\lim_{z \rightarrow -\infty} \frac{z}{z-\alpha_n^6} = 1$$

exists uniformly in n , the former limit can be computed term by term (Knopp 1928, p. 338). Therefore

$$\lim_{z \rightarrow -\infty} \psi(z) = \sum_{n=1}^{\infty} \frac{1}{\alpha_n^2}.$$

But (Watson 1944, p. 502)

$$\sum_{n=1}^{\infty} \frac{1}{\alpha_{l,n}^2} = \frac{1}{2(2l+3)}. \quad (36)$$

Thus

$$\lim_{z \rightarrow -\infty} \psi_l(z) = \frac{1}{2(2l+3)}. \quad (37)$$

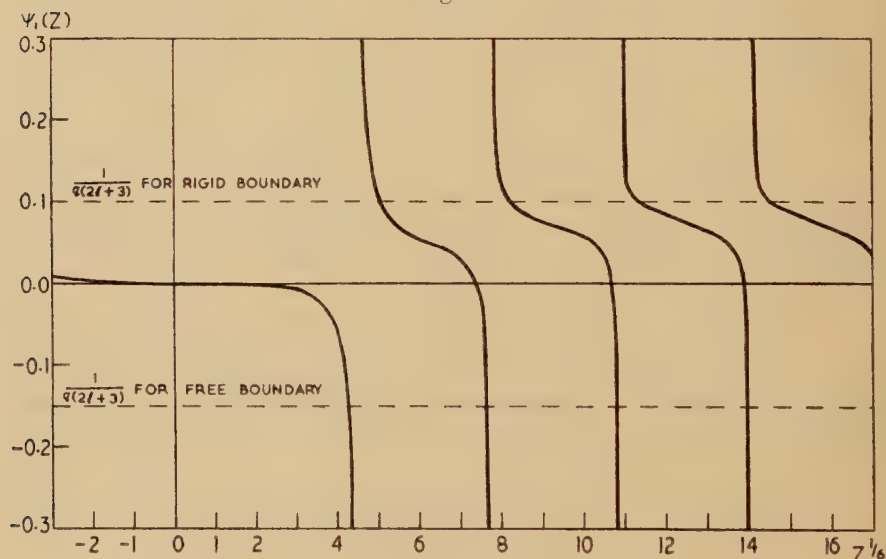
The value of this limit is precisely the value of $q(2l+1)$ for the rigid boundary problem. Therefore (35) and (37) together imply that in both the free and the rigid boundary problems, all real roots of (31) are positive.

Since

$$\psi(re^{i\theta}) = \sum_{n=1}^{\infty} \frac{r(r - \alpha_n^6 \cos \theta)}{\alpha_n^2 |r - \alpha_n^6 e^{-i\theta}|^2} - i \sin \theta \sum_{n=1}^{\infty} \frac{r \alpha_n^4}{|r - \alpha_n^6 e^{-i\theta}|^2}, \quad (38)$$

eqn. (32) can have only real roots. It is thus possible to conclude that all the roots of (32) and hence all the eigenvalues $C'_{l,m}$ are positive real numbers. In the free boundary problem there is one extra root of (32), between 0 and α_1^6 , to which there corresponds no root of the rigid boundary problem. In what follows, the root of the free boundary equation between $\alpha_{l,N}^6$ and $\alpha_{l,N+1}^6$ will be called $r_{l,N+1}$. The root of the rigid boundary equation which lies between $\alpha_{l,N}^6$ and $\alpha_{l,N+1}^6$ will be called $r_{l,N}$. Thus in both problems the roots are $r_{l,1} < r_{l,2} < r_{l,3} < \dots$.

Fig. 1



It is clear from what has been said that for a fixed l , if $r_{l,n}$ is approximated by $\alpha_{l,n}^6$, the percentage error thus committed approaches 0 as n approaches ∞ . Watson (1944, p. 506) shows that $\alpha_{l,n}$ is asymptotically equal to $(n + l/2 + \lambda_l)\pi$ for large n , where λ_l is an integer which depends only on l . Thus for large n , $r_{l,n}$ is asymptotically equal to $(n + l/2 + \lambda_l)^6 \pi^6$.

Another obvious question is the behaviour of $r_{l,n}$ for fixed n as $l \rightarrow \infty$. We shall consider only $r_{l,1}$. Chandrasekhar shows that

$$r_{l,1} \leq \frac{\alpha_{l,1}^7 J'_{l+1/2}(\alpha_{l,1})}{\alpha_{l,1} J_{l+1/2}(\alpha_{l,1}) + q J_{l+5/2}(\alpha_{l,1})}. \quad (39) \quad (I \ 54))$$

By means of the well-known recursion relations among Bessel functions (Watson 1944, p. 45) this inequality can be rewritten

$$\frac{1}{\alpha_{l,1}^6} \left(1 - \frac{q(2l+3)}{\alpha_{l,1}^2} \right) \leq \frac{1}{r_{l,1}}. \quad (40)$$

For the rigid boundary problem $r_{l,1} > \alpha_{l,1}^6$, so

$$\frac{1}{\alpha_{l,1}^6} \left(1 - \frac{q(2l+3)}{\alpha_{l,1}^2} \right) \leq \frac{1}{r_{l,1}} \leq \frac{1}{\alpha_{l,1}^6} \quad . \quad . \quad . \quad (41)$$

for rigid boundaries.

Watson (1944, p. 485) shows that

$$\alpha_{l,1} > l + \frac{1}{2}. \quad . \quad . \quad . \quad (42)$$

Then (41) implies

$$1 - \frac{2(2l+3)}{(l+\frac{1}{2})^2} \leq \frac{\alpha_{l,1}^6}{r_{l,1}} \leq 1 \quad . \quad . \quad . \quad (43)$$

for rigid boundaries. This means that as $l \rightarrow \infty$, $r_{l,1}$ is asymptotically equal to $\alpha_{l,1}^6$ and therefore to $(l+\frac{1}{2})^6$ (Watson 1944, p. 521).

For the free boundary problem more care is required. From (33),

$$\psi(z) = - \sum_{n=1}^{\infty} \frac{z}{\alpha_n^8 (1 - z/\alpha_n^6)}. \quad . \quad . \quad . \quad (44)$$

If $|z| < \alpha_1^6$,

$$\psi(z) = - \sum_{n=1}^{\infty} \frac{z}{\alpha_n^8} \sum_{k=0}^{\infty} \left(\frac{z}{\alpha_n^6} \right)^k = - \sum_{n=1}^{\infty} \sum_{k=1}^{\infty} \frac{z^k}{\alpha_n^{6k+2}}. \quad . \quad . \quad (45)$$

The Weierstrass double sum theorem (Copson 1935, p. 97) then ensures that the order of summation can be reversed, so, if $|z| < \alpha_1^6$,

$$\psi(z) = - \sum_{k=1}^{\infty} z^k \sigma_k, \quad . \quad . \quad . \quad (46)$$

where

$$\sigma_k = \sum_{n=1}^{\infty} \frac{1}{\alpha_n^{6k+2}}. \quad . \quad . \quad . \quad (47)$$

Clearly,

$$-\psi(z) \leq \sigma_1 z \sum_{k=0}^{\infty} \left(\frac{z}{\alpha_1^6} \right)^k = \frac{\alpha_1^6 \sigma_1 z}{\alpha_1^6 - z}. \quad . \quad . \quad (48)$$

Therefore the lowest root, $r_{l,1}$, of (32) is larger than the root z of

$$-\frac{\alpha_1^6 \sigma_1 z}{\alpha_1^6 - z} = -\frac{(2l+1)}{4(2l+3)}. \quad . \quad . \quad . \quad (49)$$

The root of (49) is

$$z = \frac{\alpha_1^6}{\left(1 + \frac{4(2l+3)}{(2l+1)} \alpha_1^6 \sigma_1 \right)}. \quad . \quad . \quad (50)$$

Combining this with (40),

$$\frac{1}{\alpha_{l,1}^6} \left(1 + \frac{4(2l+3)}{(2l+1)\alpha_{l,1}^2} \right) \leq \frac{1}{r_{l,1}} \leq \frac{1}{\alpha_{l,1}^6} \left(1 + \frac{(2l+1)}{4(2l+3)} \alpha_1^6 \sigma_1 \right) \quad (51)$$

for the free boundary problem. Now (51) implies that for the free boundary problem $r_{l,1}$ is asymptotically equal to $\alpha_{l,1}^6$ as $l \rightarrow \infty$, with a per cent error which behaves like $1/l$ as $l \rightarrow \infty$. To see this, use the fact that

$\alpha_{n+1} - \alpha_n$ is a decreasing function of n (Watson 1944, p. 515) which approaches π as n approaches ∞ (Watson 1944, p. 506). Then

$$\begin{aligned}\sigma_1 &= \sum_{n=1}^{\infty} \frac{1}{\alpha_n^8} \leq \frac{1}{\alpha_1^8} + \sum_{n=1}^{\infty} \frac{1}{(\alpha_1 + n\pi)^8} \leq \frac{1}{\alpha_1^8} + \int_{1/2}^{\infty} \frac{dx}{(\alpha_1 + \pi x)^8} \\ &= \frac{1}{\alpha_1^8} + \frac{1}{7\pi(\alpha_1 + \frac{1}{2}\pi)^7}.\end{aligned}$$

Hence

$$\sigma_1 \alpha_1^6 \leq \frac{1}{\alpha_1^2} + \frac{\alpha_1^6}{7\pi(\alpha_1 + \frac{1}{2}\pi)^7} \leq \frac{1}{\alpha_1^2} + \frac{1}{7\pi\alpha_1}. \quad (52)$$

Since $\alpha_{l,1} > l + \frac{1}{2}$, the assertion is established.

One remaining general remark about the distribution of the eigenvalues is that among all eigenvalues of the problem specified by eqns. (1) through (5) or (6), the smallest is $C_{1,1}$. This assertion is of some interest since the lowest of the $C_{l,n}$ is presumably the Rayleigh number at which convection will begin in an initially cool sphere which begins to warm up internally; and the lowest $C_{l,n}$ is also the Rayleigh number at which convection will cease in a sphere of fluid losing heat through its surface.

Clearly $C_{l,m} < C_{l,m+1}$ since $r_{l,m} < r_{l,m+1}$, and by eqn. (34) $C_{l,m} = r_{l,m}/l(l+1)$. There remains only to show that $C_{1,1} < C_{l,1}$ if $l > 1$. We shall shortly compute $C_{1,1}$ for both types of boundaries, after which this remaining task is easily accomplished by means of relations (41) and (42) for the rigid boundary problem and by means of relations (51) and (52) for the free boundary problem.

§ 5. COMPUTING THE EIGENVALUES AS ROOTS OF A CUBIC POLYNOMIAL

The definition (33) implies that if $\alpha_N^6 < z < \alpha_{N+1}^6$ then

$$\begin{aligned}&\sum_{n=1}^{N-1} \frac{\alpha_{N+1}^6}{\alpha_n^2(\alpha_{N+1}^6 - \alpha_n^6)} + \frac{z}{\alpha_N^2(z - \alpha_N^6)} + \frac{z}{\alpha_{N+1}^2(z - \alpha_{N+1}^6)} \\ &- z \sum_{n=N+2}^{\infty} \frac{1}{\alpha_n^2(\alpha_n^6 - \alpha_{N+1}^6)} \leq \psi(z) \leq \sum_{n=1}^{N-1} \frac{\alpha_N^6}{\alpha_n^2(\alpha_N^6 - \alpha_n^6)} + \frac{z}{\alpha_N^2(z - \alpha_N^6)} \\ &+ \frac{z}{\alpha_{N+1}^2(z - \alpha_{N+1}^6)} - z \sum_{n=N+2}^{\infty} \frac{1}{\alpha_n^2(\alpha_n^6 - \alpha_N^6)}. \quad (53)\end{aligned}$$

Since all three functions compared in (53) are monotonic decreasing with z except across their poles, the root of (32) which lies between α_N^6 and α_{N+1}^6 is bracketed between the middle roots of the two cubic polynomial equations generated by setting the two extreme terms in (53) equal to $[q(2l+3)]^{-1}$. The distance between these two middle roots of the two cubics increases like N^4 as N increases. Since $r_{l,N}$ increases like N^6 , the estimate of $r_{l,N}$ derived from (53) has a per cent error which decreases like N^{-2} as N increases.

However, computation of the coefficients of the two cubics generated from (53) is quite laborious, and $r_{l,N}$ can be computed more simply if l is not too large and $n=1$ or 2. It is necessary to observe that

$$\psi(z) = \frac{z}{\alpha_1^2(z - \alpha_1^6)} + \frac{z}{\alpha_2^2(z - \alpha_2^6)} - z(1+Q) \left(\sum_{n=3}^{\infty} \frac{1}{\alpha_n^8} \right) \quad (54)$$

From these tables it is clear that the eigenvalues $C'_{l,2}$ are too high to appear before all $C'_{l,1}$ convection patterns have begun for l at least up to 6. From Chandrasekhar's tables 1 and 2 it appears that, if the boundary is free, convection patterns $C'_{l,1}$ for l up to 11 set in before any $C'_{l,2}$ begins; and, if the boundary is rigid, the $C'_{l,1}$ patterns for l up to 8 set in before any $C'_{l,2}$ pattern appears. It should be noted, however, that an attempt to use tables 1 and 2 to predict when various patterns of convection begin meets with the difficulty that the linearized Navier-Stokes equation no longer describes the flow once any convection pattern has acquired a finite amplitude.

From the tables 1 and 2 observe that Chandrasekhar's variational values for $C'_{l,1}$ for l from 1 through 6 are accurate to one part in 10^4 in the free boundary problem and to three parts in 10^3 in the rigid boundary problem. Note also that $C'_{1,2} > C'_{2,2}$.

§ 6. COMPUTING THE EIGENVALUES WITH ARBITRARY ACCURACY

Very accurate values for the roots of (32) can be computed rather quickly by Euler's method for computing the zeros of $J_0(z)$ (Watson 1944, p. 500). The advantage of this technique is that, like Newton's method, it can be carried to unlimited accuracy, while less computation is required than in Newton's method.

Since the infinite product $\prod_{n=1}^{\infty} (1 - z/\alpha_{l,n}^6)$ converges absolutely and, in any bounded region of the complex plane, uniformly to an analytic function whose only zeros are simple zeros at $z = \alpha_n^6$, we can define

$$g_l(z) = \left[1 - \frac{1}{q(2l+3)} \psi_l(z) \right] \prod_{n=1}^{\infty} \left(1 - \frac{z}{\alpha_{l,n}^6} \right). \quad (58)$$

The function $g(z)$ is analytic in the whole complex plane, $g(0)=1$, and the roots $r_{l,n}$ of (32) are the zeros of $g(z)$. Using the three facts that $|\alpha_{l,n+1} - \alpha_{l,n}| > \pi$, $\alpha_{l,1} > l + \frac{1}{2}$, and that for each l there is a real number λ_l such that for sufficiently large n $\pi(n + \frac{1}{2}l + \lambda_l) < \alpha_{l,n} < \pi(n + \frac{1}{2}l + \lambda_l + \frac{1}{4})$ (Watson 1944, pp. 506, 485 and 492 respectively) it can be shown that if

$$M_{l,n} = \sup \left\{ \left| \frac{g'_l(z)}{g_l(z)} \right| : |z| = \alpha_{l,n}^6 \right\} \quad (59)$$

then

$$\lim_{n \rightarrow \infty} M_{l,n} = 0. \quad (60)$$

A slight modification of the theorem on p. 137 of Whittaker and Watson (1952) then shows that

$$g_l(z) = \prod_{n=1}^{\infty} \left(1 - \frac{z}{r_{l,n}} \right). \quad (61)$$

Therefore, if

$$g_l(z) = \sum_{k=0}^{\infty} g_{l,k} z^k, \quad (62)$$

it follows that

$$g_{l,k}=(-1)^k \sum_{n_1 < n_2 < \dots < n_k} \frac{1}{r_{l,n_1} r_{l,n_2} \dots r_{l,n_k}}, \quad \dots \quad (63)$$

and Graeffe's technique of root squaring may be applied to the power series for $g(z)$. The procedure for computing zeros of (58) is to compute the coefficients in the power series for $g(z)$ from (58) and (33), and then to compute (Graeffe 1837, p. 17 ff.) the power series for the functions whose roots are respectively the squares, fourth powers, eighth powers, etc., of the roots of $g(z)$. In this connection it is necessary and not difficult to show that the power series obtained by applying Graeffe's algorithm to $g(z)$ is simply the infinite product in (61) with $r_{l,n}$ replaced by $r_{l,n}^2$. If

$$g_l^{(n)}(z) = \sum_{k=0}^{\infty} g_{l,k}^{(n)} z^k \quad \dots \quad (64)$$

is the power series obtained from (62) by squaring its roots n times, then

$$g_{l,1}^{(n)} = - \sum_{m=1}^{\infty} (r_{l,m})^{-(2^n)}. \quad \dots \quad (65)$$

Since $0 < r_{l,1} < r_{l,2} < \dots$ and $\sum_{m=1}^{\infty} \frac{1}{r_{l,m}}$

converges, therefore if n is large enough it is approximately true that

$$g_{l,1}^{(n)} = -(r_{l,1})^{-(2^n)}. \quad \dots \quad (66)$$

For $l=1$, the lowest root $r_{1,1}$ of (58) can be computed to eight places by taking $n=2$; that is, by two root-squaring processes. The results are

$$C_{1,1} = 3091.1927 \pm 0.0001 \text{ for a free boundary}$$

and

$$C_{1,1} = 8040.0469 \pm 0.0003 \text{ for a rigid boundary.}$$

§ 7. CONVECTION IN A HORIZONTAL LAYER OF FLUID

The method described in this paper can be applied to the classical Rayleigh-Benard convection problem in a horizontal layer of viscous fluid heated below (Lord Rayleigh 1916). Supposing that the two boundaries of the layer are at $z = \pm \frac{1}{2}$, the eigenvalue problem to be solved consists of the equation

$$\left(\frac{d^2}{dz^2} - a^2 \right)^3 W = -\lambda W \quad \dots \quad (67)$$

together with the boundary conditions

$$W = 0 \text{ at } z = \pm \frac{1}{2}, \quad \dots \quad (68)$$

$$\left(\frac{d^2}{dz^2} - a^2 \right)^2 W = 0 \text{ at } z = \pm \frac{1}{2}, \quad \dots \quad (69)$$

and

$$\left. \begin{aligned} W' &= 0 \text{ at a free boundary,} \\ W'' &= 0 \text{ at a rigid boundary.} \end{aligned} \right\} \quad \dots \quad (70)$$

or

Here $\lambda_1 a^2$ is the Rayleigh number, and ' a ' is the dimensionless horizontal wave number in eqn. (34) of Chandrasekhar (1953 a). For each positive number a , there is a critical Rayleigh number, $\lambda(a)$, at which convection will set in whose horizontal wave number will be a . The minimum value of $\lambda(a)$ as a varies from 0 to ∞ is the critical Rayleigh number at which convection will begin in the horizontal layer. For each a , $\lambda(a)$ is determined as the eigenvalue in (67). To compute $\lambda(a)$, let

$$F = \left(\frac{d^2}{dz^2} - a^2 \right)^2 W. \quad . \quad . \quad . \quad . \quad . \quad (71)$$

Define

$$\mu_n = (2n+1)\pi, \quad \nu_n = 2n\pi \quad . \quad . \quad . \quad . \quad (72)$$

and expand F in Fourier series on $[-\frac{1}{2}, \frac{1}{2}]$:

$$F = \sum_{n=0}^{\infty} A_n \cos \mu_n z + \sum_{n=1}^{\infty} B_n \sin \nu_n z. \quad . \quad . \quad . \quad . \quad (73)$$

Then integrating (71) gives

$$W = \sum_{n=0}^{\infty} \frac{A_n}{(\mu_n^2 + a^2)^2} \cos \mu_n z + \sum_{n=0}^{\infty} \frac{B_n}{(\nu_n^2 + a^2)^2} \sin \nu_n z + A \cosh az + Bz \sinh az + C \sinh az + Dz \cosh az. \quad . \quad . \quad (74)$$

But (67), expressed in terms of F , is

$$\left(\frac{d^2}{dz^2} - a^2 \right) F = -\lambda W. \quad . \quad . \quad . \quad . \quad . \quad (75)$$

Substituting the expression for W given by (74) in (75) and integrating, one obtains

$$\begin{aligned} -\frac{F}{\lambda} = & - \sum_{n=0}^{\infty} \frac{A_n}{(\mu_n^2 + a^2)^3} \cos \mu_n z - \sum_{n=1}^{\infty} \frac{B_n}{(\nu_n^2 + a^2)^3} \sin \nu_n z + \left(\frac{A}{2a} - \frac{B}{4a^2} \right) \\ & \times z \sinh az + \frac{B}{4a} z^2 \cosh az + \left(\frac{C}{2a} - \frac{D}{4a^2} \right) z \cosh az + \frac{D}{4a} z^2 \sinh az \\ & + E \sinh az + G \cosh az. \quad . \quad . \quad . \quad . \quad . \quad (76) \end{aligned}$$

Boundary conditions (68) and (69) imply that there exist constants P and Q such that

$$\left. \begin{aligned} A &= P \sinh \tfrac{1}{2}a, \\ B &= -2P \cosh \tfrac{1}{2}a, \\ G &= \left(\frac{a - a \sinh^2 \tfrac{1}{2}a - 2 \sinh \tfrac{1}{2}a \cosh \tfrac{1}{2}a}{8a^2 \cosh \tfrac{1}{2}a} \right) P, \end{aligned} \right\} \quad . \quad . \quad (77)$$

and

$$\left. \begin{aligned} C &= Q \cosh \tfrac{1}{2}a, \\ D &= -2Q \sinh \tfrac{1}{2}a, \\ E &= - \frac{(a + a \cosh^2 \tfrac{1}{2}a + 2 \sinh \tfrac{1}{2}a \cosh \tfrac{1}{2}a)}{8a^2 \sinh \tfrac{1}{2}a} Q. \end{aligned} \right\} \quad . \quad . \quad (78)$$

Inserting (73) on the left of (76) and taking Fourier coefficients,

$$A_n = P \left[16a \cosh^2 \frac{a}{2} \left(\frac{(-1)^n \mu_n \lambda}{(\mu_n^2 + a^2)^3 - \lambda} \right) \right], \quad . \quad . \quad . \quad (79)$$

and

$$B_n = Q \left[16a \sinh^2 \frac{a}{2} \left(\frac{(-1)^n \nu_n \lambda}{\lambda - (\nu_n^2 + a^2)^3} \right) \right]. \quad . \quad . \quad . \quad (80)$$

Now boundary conditions (70) can be applied at the two boundaries to determine λ and hence A_n and B_n . For example, if both boundaries are rigid, differentiating (74) and setting the result equal to 0 at $z = \pm \frac{1}{2}$ gives

$$P = -\frac{1}{a + \sinh a} \sum_{n=0}^{\infty} \frac{\mu_n A_n (-1)^n}{(\mu_n^2 + a^2)^2}, \quad . \quad . \quad . \quad (81)$$

and

$$Q = -\frac{1}{a - \sinh a} \sum_{n=1}^{\infty} \frac{\nu_n B_n (-1)^n}{(\nu_n^2 + a^2)^2}. \quad . \quad . \quad . \quad (82)$$

If (79) and (80) be substituted in (81) and (82) the result is

$$P \left[\frac{a + \sinh a}{16a \cosh^2 \frac{1}{2}a} + \sum_{n=0}^{\infty} \frac{\lambda \mu_n^2}{(\mu_n^2 + a^2)^2 [(\mu_n^2 + a^2)^3 - \lambda]} \right] = 0, \quad . \quad (83)$$

and

$$Q \left[\frac{a - \sinh a}{16a \sinh^2 \frac{1}{2}a} - \sum_{n=1}^{\infty} \frac{\lambda \nu_n^2}{(\nu_n^2 + a^2)^2 [(\nu_n^2 + a^2)^3 - \lambda]} \right] = 0. \quad . \quad (84)$$

If $P \neq 0$, (83) is an equation for λ whose solutions, by means of (79), produce even eigenfunctions, so that $Q = 0$. If $Q \neq 0$, (84) gives λ , which produces odd eigenfunctions, so that $P = 0$.

The case where one boundary is rigid and the other free does not give rise to eigenfunctions with a definite parity, so the characteristic equation for λ , analogous to (83) and (84), is more complicated, although the principle is the same. Finally, if both boundaries are free, Rayleigh's original solution (Rayleigh 1916) results from the above analysis.

It would not be useful to reproduce here the eigenvalues one actually obtains from the above computations, since the classical Rayleigh problem for both rigid and free boundaries has already been solved by a variety of methods, and the results are available in the literature. See, for example, Low (1929) or Pellew and Southwell (1940).

§ 8. MATHEMATICAL SUMMARY

Because there appears to be a class of problems amenable to the attack used in this paper, we shall describe the method in general terms. The difficulties produced by singular points will be ignored in this discussion.

Let $\mathfrak{D}$ be a linear differential operator of order d with continuous coefficients on the interval $0 \leq x \leq 1$. Let $\mathfrak{B}$ be a class of d linear homogeneous boundary conditions on a function and its first $d-1$ derivatives at $x=0$ and $x=1$. The eigenvalue problem to be solved for eigenvalues λ and eigenfunctions W is given by the two requirements that

$$\mathfrak{D}W = \lambda W \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (85)$$

and that W satisfies the boundary conditions $\mathfrak{B}$ (86)

The solution consists of three steps. The first is to find a set of functions ϕ_n such that any continuous function W satisfying the conditions $\mathfrak{B}$ can be expanded in a series

$$W = \sum_{n=1}^{\infty} A_n \phi_n \quad (87)$$

with constant coefficients A_n which is uniformly convergent on $[0, 1]$.

The second step consists in substituting (87) on the right of (85) and integrating (85) term by term. The result of the substitution is

$$\mathfrak{B}W = \lambda \sum_{n=1}^{\infty} A_n \phi_n. \quad (88)$$

To perform the integration one must first find the functions $\psi_n(x)$ which satisfy the two conditions

$$\mathfrak{B}\psi_n(x) = \phi_n(x) \quad \text{on} \quad 0 \leq x \leq 1 \quad (89)$$

and

$$\psi_n \text{ and its first } d-1 \text{ derivatives are zero at } x=0.* \quad . . . (90)$$

Then one must use the fact that because (87) is uniformly convergent, (88) has the particular solution

$$\bar{W} = \sum_{n=1}^{\infty} \lambda A_n \psi_n(x), \quad (91)$$

where the series is uniformly convergent on $[0, 1]$. This can be shown by a slight modification of the argument on p. 72 of Ince (1944). Finally, one must compute the general solution Q of

$$\mathfrak{B}Q = 0. \quad (92)$$

This solution Q will have the form

$$Q(x) = \sum_{j=1}^d Q_j q_j(x) \quad (93)$$

where the q_j are d linearly independent solutions of (92) and the Q_j are arbitrary constants. Then the result of step 2 is the equation

$$\sum_{n=1}^{\infty} A_n (\phi_n - \lambda \psi_n) = Q. \quad (94)$$

Step 3 requires finding the dual basis for the functions $\phi_n - \lambda \psi_n$, that is, the set of functions $\zeta_n(x, \lambda)$ such that

$$\int_0^1 [\phi_n(x) - \lambda \psi_n(x)] \zeta_m(x, \lambda) dx = \delta_{mn}. \quad (95)$$

* This condition can, in many particular cases, be replaced by less restrictive conditions which still ensure the uniform convergence of (91). Clearly 0 can be replaced here by any point in $[0, 1]$.

If $k_{jm}(\lambda)$ is defined as

$$k_{jm}(\lambda) = \int_0^1 \zeta_m(x, \lambda) q_j(x) dx, \quad (96)$$

then multiplying (94) on both sides by ζ_m and integrating from 0 to 1 gives

$$A_m = \sum_{j=1}^d Q_j k_{jm}(\lambda). \quad (97)$$

If these expressions (97) for A_m are substituted in (87), and then the d boundary conditions $\mathfrak{B}$ are applied to W , one obtains d linear homogeneous equations in the constants Q_j , the coefficients of which are infinite series involving λ . Setting the d by d determinant of this system equal to 0 produces a characteristic equation for the eigenvalues λ . Any solution of this equation can be substituted in (97) to give the A_m and hence, via (87), the eigenfunction W appropriate to the given eigenvalue.

The methods by which these three steps can be carried out most conveniently will depend on the nature of the problem at hand. In both problems considered in this paper, $\mathfrak{B}$ is of the form

$$\mathfrak{B} = \mathfrak{D}_1 \mathfrak{D}_2 \dots \mathfrak{D}_p \quad (98)$$

where $\mathfrak{D}_i$ is a linear differential operator of order d_i ; and the set $\mathfrak{B}$ of boundary conditions contains a subclass $\mathfrak{C}$ of d_1 boundary conditions on a function and its first $d_1 - 1$ derivatives. The class ϕ_n can be taken as the class of eigenfunctions of the eigenvalue problem

$$\mathfrak{D}_1 \phi = \alpha \phi \quad (99)$$

and ϕ satisfies boundary conditions $\mathfrak{C}$ (100)

Then the uniform convergence of (87), at least for the second order operators $\mathfrak{D}_1$ considered in this paper, is assured by the fact that W must satisfy boundary conditions $\mathfrak{C}$. Furthermore, in both the problems of this paper, there are numbers $\alpha_{n,i}$ such that

$$\mathfrak{D}_i \phi_n = \alpha_{n,i} \phi_n, \quad (101)$$

so that we can take

$$\psi_n = \frac{\phi_n}{\alpha_{n,1} \dots \alpha_{n,p}}. \quad (102)$$

Finally, in both problems considered in this paper, (99) and (100) form a self-adjoint boundary value problem so that we can take ζ_n to be ϕ_n except for a normalization constant.

In case the problem under consideration has all the characteristics described in the preceding paragraph except the self-adjointness of (99) and (100), one can still take ψ_n in the form (102), but ζ_n is obtained as an

eigenfunction of the eigenvalue problem adjoint to (99) and (100) (Ince 1944, pp. 210–212).

In general, if steps one and two can be carried out, step three can always be performed in principle as follows*: let $\theta_n(x)$ be an orthonormal basis constructed from the $\phi_n - \lambda\psi_n$ by the Gramm–Schmidt process, so that one has

$$\theta_i(x, \lambda) = \sum_{j=1}^i c_{ij}(\lambda)(\phi_j - \lambda\psi_j) \quad . \quad . \quad . \quad (103)$$

where $c_{ii}(\lambda) = 0$ if $i < j$. Take $\zeta_n = \sum_{m=1}^{\infty} c_{mn}(\lambda)\theta_m$. Then ζ_n is the required dual basis.

§ 9. CONCLUSION

The principal result of this paper can be briefly described. Suppose one seeks the eigenfunctions W and eigenvalues λ of an eigenvalue problem of high order d , of the form given by eqns. (85) and (86), where the boundary conditions $\mathfrak{B}$ are d linear homogeneous equations in W and its first $d-1$ derivatives at 0 and 1. Whenever there is a class of functions ϕ_n such that any W satisfying the boundary conditions $\mathfrak{B}$ can be expanded as a uniformly convergent series of such functions ϕ_n , it is possible to obtain a characteristic equation for the eigenvalue λ in the form of a d by d determinant whose entries are infinite series in λ .

This method is of interest only when, as in the problem of the fluid sphere heated from within, it gives a characteristic equation which is easier to solve than the characteristic equation obtained by straightforward methods. For Rayleigh's classical problem of convection in a horizontal layer of fluid nothing new is obtained, but in the problem of the fluid sphere some new information about the higher radial convection patterns with a given angular symmetry can be deduced. Also, in the latter problem the method presented here can be used as a check on the accuracy of Chandrasekhar's more convenient variational method. It is found that the variational method is quite accurate: two approximations are sufficient to give three or four significant figures in the eigenvalues, depending on the boundary conditions.

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* This observation is due to Professor S. Chandrasekhar.

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Note added in proof.—The author finds that equations (83) and (84) were derived in a different way by Jeffreys, *Proc. Roy. Soc. A*, **118**, 195. See equation (13) on page 200.

CXLIII. *The Influence of Retardation on
the London-van der Waals Forces*

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ABSTRACT

A calculation is made of the effect of including the retarded potential term of quantum electrodynamics when evaluating the van der Waals interaction between two atoms. In disagreement with the conclusions of Casimir and Polder the interaction energy at large distances is found to vary as R^{-3} .

§ 1. INTRODUCTION

THE retarded interaction of a pair of atoms was the subject of a paper by Casimir and Polder (1948) (hereafter C-P). These authors obtained the energy of interaction by means of a perturbation method using the electronic charge e as a perturbation parameter. The energy is $\sim e^4$. In the course of their calculation the unavoidable infinities of energy make their appearance but are shown to be of no significance since they are independent of the separation of the atoms. The standard result, obtained by Eisenschitz and London (1930) considering only the electrostatic interaction, is that the interaction energy is attractive and $\propto R^{-6}$. C-P find that this is modified, at large distances, to an attractive energy $\propto R^{-7}$. This result has been widely quoted (e.g. Verwey and Overbeek 1948).

A closer inspection shows that the C-P result cannot be regarded as a direct consequence of the principles of quantum field theory but depends on the particular method of approximation used by them. Starting from two atoms and the field of radiation *in vacuo* they first obtain the distortion ($\sim e^2$) of the radiation field due to the presence of one of the atoms. This distorted field and the second atom in its ground state are then used as the zero approximation for a second stage of perturbation which, being carried to terms $\sim e^2$, yields energies $\sim e^4$ as required.

There is no necessity for proceeding in this way in which there is an artificial asymmetry between the atoms. As an alternative the original unperturbed state can be corrected to fourth order in the energy in a completely symmetric way by systematic application of the Rayleigh-Schrödinger perturbation theory.

* Communicated by the Author,

It is, moreover, no foregone conclusion that the two methods should yield the same result. It is well known that the solution of differential equations by the method of successive approximations, in particular by expansion in powers of a small parameter, does not lead in general to unique results. An instructive example was recently given by Kronig and Bruijsten (1950) who showed that an apparently innocuous problem in thermal conduction may become insoluble if the zero approximation is not carefully chosen.

In the present problem it is quite likely that the two methods should not agree. The zero approximation of the conventional perturbation theory is a non-degenerate state. According to the general formulae of the Schrödinger theory (see for example Condon and Shortley 1951, pp. 30-35) there can be no resonance denominators in the matrix elements in any degree of approximation. In contrast the matrix elements given by C-P show resonances due to the degeneracy of the distorted field of radiation.

In this paper the conventional perturbation theory for the above system has been carried through with a result differing markedly from that of C-P. Since these authors assert that they have checked their unconventional method against the ordinary theory, the intermediate steps in the present calculation are presented in some detail so that there can be no doubt that this particular assertion of C-P is wrong.

Instead of the reduction of the attraction at large distances as found by C-P it is now found to be increased by the retardation, although the increase is likely to be insufficient for direct observation. The indirect consequences of the increase are, however, by no means insignificant and are the subject of a further investigation now in progress.

The question remains whether the present conventional perturbation method or the asymmetric approach of C-P is the correct one. Subject to the reservation that, at the present stage of quantum field theory, a question of this kind may be premature, it cannot reasonably be doubted that the present approach is to be preferred because of the lack of singularities in the intermediate stages.

§ 2. METHOD OF CALCULATION

The unperturbed state of the system consists of atoms A and B and the radiation field all independently occupying their ground states and is represented by the wave function

$$\Psi_0 = \psi_0^A \psi_0^B \psi_0^F. \quad (1)$$

The perturbation is represented by the addition to the Hamiltonian of the term

$$H' = Q + \sum_{A,B} \left(-\frac{e}{mc} \mathbf{p} \cdot \mathbf{A} + \frac{e^2}{2mc^2} A^2 \right) \quad (2)$$

where

$$Q = \frac{\mathbf{q}_A \cdot \mathbf{q}_B}{R^3} - 3 \frac{\mathbf{q}_A \cdot \mathbf{R} \mathbf{q}_B \cdot \mathbf{R}}{R^5} \quad (3)$$

is the electrostatic interaction term whose effect is determined in the standard van der Waals calculation

$$imck_r \mathbf{q}_A = -e \sum_A \mathbf{p}. \quad . \quad . \quad . \quad . \quad . \quad (4)$$

$\sum_A$ denotes summation over all the electrons of atom A.

$$\hbar ck_r = E_r - E_0. \quad . \quad . \quad . \quad . \quad . \quad (5)$$

E_0 , E_r are the ground and excited state energies of an atom.

$$\mathbf{A} = \sum_{\mathbf{k}, \lambda} e C_{\mathbf{k}} \mathbf{e}(\mathbf{k}, \lambda) \{ A_{\mathbf{k}, \lambda}^- \exp(i\mathbf{k} \cdot \mathbf{r}) + A_{\mathbf{k}, \lambda}^+ \exp(-i\mathbf{k} \cdot \mathbf{r}) \}. \quad . \quad (6)$$

$A_{\mathbf{k}, \lambda}^-$ and $A_{\mathbf{k}, \lambda}^+$ are respectively the annihilation and creation operators of quantum electrodynamics with the properties

$$\left. \begin{aligned} A_{\mathbf{k}, \lambda}^- \psi^F(\dots N_{\mathbf{k}, \lambda} \dots) &= \sqrt{N} \psi^F(\dots (N-1)_{\mathbf{k}, \lambda} \dots) \\ A_{\mathbf{k}, \lambda}^+ \psi^F(\dots N_{\mathbf{k}, \lambda} \dots) &= \sqrt{N+1} \psi^F(\dots (N+1)_{\mathbf{k}, \lambda} \dots) \end{aligned} \right\} \quad . \quad (7)$$

$C_{\mathbf{k}} = (2\pi\hbar/\omega L^3)^{1/2}$ (it having been assumed for normalization purposes that the system is enclosed in a cubical box of side L where $L \rightarrow \infty$ finally).

$\mathbf{e}(\mathbf{k}, \lambda)$ is the unit vector representing the polarization of the photon whose wave vector is $\mathbf{k}$. λ denotes the two polarization possibilities corresponding to each value of $\mathbf{k}$.

It is convenient to regard the electronic charge e as the perturbation parameter and the full perturbed Hamiltonian may be written

$$H_0' = H_0 + H' = H_0 + eH_1 + e^2H_2 \quad . \quad . \quad . \quad . \quad . \quad (8)$$

where

$$eH_1 = - \sum_{A, B} \frac{e}{mc} \mathbf{p} \cdot \mathbf{A}, \quad . \quad . \quad . \quad . \quad . \quad (9)$$

$$e^2H_2 = Q + \sum_{A, B} \frac{e^2}{2mc^2} A^2. \quad . \quad . \quad . \quad . \quad . \quad (10)$$

(N.B. Q contains the factor e^2 .)

The perturbed ground state wave function is assumed to be of the form

$$\Psi_0' = \Psi_0 + e \sum_{n=(r, s, t)} a_{rst} \psi_r^A \psi_s^B \psi_t^F + e^2 \sum_n b_{rst} \psi_r^A \psi_s^B \psi_t^F + \dots \quad (11)$$

and the perturbed ground state energy may similarly be expressed in the form of a series

$$E_0' = E_0 + eE_0^{(1)} + e^2E_0^{(2)} + \dots \quad . \quad . \quad . \quad . \quad . \quad (12)$$

where it may be shown that the first four perturbed terms are given by

$$E_0^{(1)} = (H_1)_{00}, \quad . \quad . \quad . \quad . \quad . \quad (13)$$

$$E_0^{(2)} = (H_2)_{00} + \sum_{n \neq 0} \frac{1}{E_0 - E_n} |(H_1)_{0n}|^2, \quad . \quad . \quad . \quad . \quad . \quad (14)$$

$$\begin{aligned} E_0^{(3)} = & \sum_{n \neq 0} \frac{1}{E_0 - E_n} \{ (H_2)_{0n} (H_1)_{n0} + (H_1)_{0n} (H_2)_{n0} \} \\ & + \sum_{n \neq 0} \sum_{n' \neq 0} \frac{1}{(E_0 - E_n)(E_0 - E_{n'})} (H_1)_{0n} (H_1)_{nn'} (H_1)_{n'0} \\ & - (H_1)_{00} \sum_{n \neq 0} \frac{1}{(E_0 - E_n)^2} |(H_1)_{0n}|^2, \quad . \quad . \quad . \quad . \quad . \quad (15) \end{aligned}$$

$$E_0^{(4)} = (a) + (b) + (c) + (d) + (e), \quad , \quad , \quad , \quad , \quad , \quad (16)$$

where

$$\begin{aligned}
 (a) &= \sum_{n \neq 0} \sum_{n' \neq 0} \sum_{n'' \neq 0} \frac{1}{(E_0 - E_n)(E_0 - E_{n'})(E_0 - E_{n''})} \\
 &\quad \times (H_1)_{0n} (H_1)_{nn'} (H_1)_{n'n''} (H_1)_{n''0}, \\
 (b) &= \sum_{n \neq 0} \sum_{n' \neq 0} \frac{1}{(E_0 - E_n)(E_0 - E_{n'})} \\
 &\quad \times \{ (H_2)_{0n} (H_1)_{nn'} (H_1)_{n'0} + (H_1)_{0n} (H_2)_{nn'} (H_1)_{n'0} + (H_1)_{0n} (H_1)_{nn'} (H_2)_{n'0} \}, \\
 (c) &= \sum_{n \neq 0} \frac{1}{E_0 - E_n} |(H_2)_{0n}|^2, \\
 (d) &= - \left\{ (H_2)_{00} + \sum_{n \neq 0} \frac{1}{E_0 - E_n} |(H_1)_{0n}|^2 \right\} \sum_{n \neq 0} \frac{1}{(E_0 - E_n)^2} |(H_1)_{0n}|^2, \\
 (e) &= (H_1)_{00}^2 \sum_{n \neq 0} \frac{1}{(E_0 - E_n)^3} |(H_1)_{0n}|^2 - 2(H_1)_{00} \sum_{n \neq 0} \sum_{n' \neq 0} \frac{(H_1)_{0n} (H_1)_{nn'} (H_1)_{n'0}}{(E_0 - E_n)^2 (E_0 - E_{n'})} \\
 &\quad - (H_1)_{00} \sum_{n \neq 0} \frac{1}{(E_0 - E_n)^2} \{ (H_1)_{0n} (H_2)_{n0} + (H_2)_{0n} (H_1)_{n0} \}.
 \end{aligned}$$

(These formulae include the effects of renormalization of the wave function and agree with those quoted in Condon and Shortley 1948.)

The following simplifying assumptions (identical with those of C-P) are made :

(a) Atoms A and B are identical and so far apart that exchange effects can be neglected.

(b) The ground states of the atoms are S states (in other cases there is little interest in the van der Waals forces).

(c) The only effective excited state is the first accessible triply degenerate P state (all other states will contribute only to a lower order of magnitude).

(d) The factor $\exp(i\mathbf{k} \cdot \mathbf{r})$ can be considered constant for the integration over the coordinate space of either atom (this is a common procedure in radiation theory and neglects quadrupole and higher order moments).

With these assumptions no difficulties are encountered in the evaluation of the various matrix elements which are found to have the following values :

$$\begin{aligned}
 e(H_1)_{00} &= 0, \quad \dots \dots \dots (17) \\
 e(H_1)_{nn'} &= \left. \begin{aligned}
 &= icC_k \{ k_{r'} \exp(i\mathbf{k}' \cdot \mathbf{R}_A) (\mathbf{q})_{0r'} \delta_{0s'} + k_{s'} \exp(i\mathbf{k}' \cdot \mathbf{R}_B) (\mathbf{q})_{0s'} \delta_{0r'} \} \cdot \mathbf{e}(\mathbf{k}, \lambda) \\
 &\quad (n = (0; 0; 0) \quad n' = (r'; s'; \mathbf{k}', \lambda')) \\
 &= -icC_k \{ k_r \exp(-i\mathbf{k} \cdot \mathbf{R}_A) (\mathbf{q})_{r0} \delta_{s0} + k_s \exp(-i\mathbf{k} \cdot \mathbf{R}_B) (\mathbf{q})_{s0} \delta_{r0} \} \cdot \mathbf{e}(\mathbf{k}, \lambda) \\
 &\quad (n = (r; s; \mathbf{k}, \lambda) \quad n' = (0; 0; 0)) \\
 &= icC_k \{ k_{r'} \exp(-i\mathbf{k} \cdot \mathbf{R}_A) (\mathbf{q})_{0r'} \delta_{0s'} + k_{s'} \exp(-i\mathbf{k} \cdot \mathbf{R}_B) (\mathbf{q})_{0s'} \delta_{0r'} \} \cdot \mathbf{e}(\mathbf{k}, \lambda) \\
 &\quad (n = (0; 0; 0; \mathbf{k}, \lambda) \quad n' = (r'; s'; 0)) \text{ etc.}
 \end{aligned} \right\} \dots \dots \dots (18)
 \end{aligned}$$

$$e^2(H_2)_{nn'} = (Q)_{nn'} + \sum_{A,B} \frac{e^2}{2mc^2} (A^2)_{nn'} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (19)$$

$$(Q)_{nn'} = \delta_{nn'} \left\{ \frac{(\mathbf{q})_{rr'} \cdot (\mathbf{q})_{ss'}}{R^3} - \frac{3(\mathbf{q})_{rr'} \cdot \mathbf{R}(\mathbf{q})_{ss'} \cdot \mathbf{R}}{R^5} \right\} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (20)$$

$$\sum_{A,B} (A^2)_{00} = 2c^2 \sum_{\mathbf{k}} C_k^2 \sum_{A,B} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (21)$$

$$\sum_{A,B} (A^2)_{0n} = 2\delta_{0r}\delta_{0s}c^2C_kC_{k'} \left\{ \exp[i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{R}_A] \sum_A + \exp[i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{R}_B] \sum_B \right\} \\ \times \sum_i e_i(\mathbf{k}, \lambda) e_i(\mathbf{k}', \lambda') \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (22)$$

$$\sum_{A,B} (A^2)_{n0} = 2\delta_{r0}\delta_{s0}c^2C_kC_{k'} \left\{ \exp[-i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{R}_A] \sum_A + \exp[-i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{R}_B] \sum_B \right\} \\ \times \sum_i e_i(\mathbf{k}, \lambda) e_i(\mathbf{k}', \lambda') \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (23)$$

(N.B. $n=(r; s; \mathbf{k}, \lambda+\mathbf{k}', \lambda')$ in (22) or (23) and a further factor $1/\sqrt{2}$ is required if $\mathbf{k}, \lambda \equiv \mathbf{k}', \lambda'$.)

$$\sum_{A,B} (A^2)_{nn'} = 2\delta_{rr'}\delta_{ss'}c^2C_kC_{k'} \left\{ \exp[i(\mathbf{k}'-\mathbf{k}) \cdot \mathbf{R}_A] \sum_A + \exp[i(\mathbf{k}'-\mathbf{k}) \cdot \mathbf{R}_B] \sum_B \right\} \\ \times \sum_i e_i(\mathbf{k}, \lambda) e_i(\mathbf{k}', \lambda') \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (24)$$

$$(n=(r; s; \mathbf{k}, \lambda) \quad n'=(r'; s'; \mathbf{k}', \lambda') \quad \mathbf{k}, \lambda \neq \mathbf{k}', \lambda'),$$

$$\sum_{A,B} (A^2)_{nn} = 2c^2 \left\{ C_k^2 + \sum_{\mathbf{k}'} C_{k'}^2 \right\} \sum_{A,B} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (25)$$

$$(n=(r; s; \mathbf{k}, \lambda)).$$

The perturbation energy ($E_0' - E_0$) contains a number of terms arising from the separate interactions between atom A and the field and between atom B and the field. These are omitted as being irrelevant to the problem of the interaction between the atoms. The interaction energy calculated by modifying the perturbation energy in this way will be termed ΔE .

Following C-P the coordinate axes are so chosen that the axis of separation of A and B lies in the z direction and the degenerate p wave functions are so selected that the corresponding dipole moments lie in the x , y and z directions respectively. In the course of the calculations summation over $\mathbf{k}$ is replaced by an integration over $\mathbf{k}$ space with the correspondence

$$\sum_{\mathbf{k}} \rightarrow \frac{L^3}{8\pi^3} \int d\mathbf{k}.$$

$$\text{The equalities} \quad \frac{e^2}{2mc^2} \sum_A = \sum_r \frac{k_r}{c\hbar} |(q^z)_{0r}|^2 = \frac{k_r}{c\hbar} |(\mathbf{q})_{0r}|^2 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (26)$$

$$\text{and} \quad \sum_{\lambda} e_i^2(\mathbf{k}, \lambda) = 1 - k_i^2/k^2 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (27)$$

are utilized and it is convenient to make the substitutions $x=kR$ and $a=k_rR$.

§ 3. RESULTS

On examining the perturbation terms in detail it is readily seen that $E_0^{(1)}$, $E_0^{(3)}$ and (e) of $E_0^{(4)}$ are all zero. In addition $E_0^{(2)}$ is eliminated by the removal of the separate (A—F) and (B—F) interaction terms. Of the terms to fourth order this leaves only (a) , (b) , (c) and (d) of $E_0^{(4)}$. These may be divided into three groups: those arising from the purely electrostatic interaction, those from the purely retarded field interaction and the mixed terms.

There is only one representative in the first group ; it comes from one half of term (c) and is given by

$$X_1 = \frac{-3 |(\mathbf{q})_{0r}|^4}{\hbar c k_r R^6}. \quad (28)$$

This is, of course, identical with the Eisenschitz–London result.

The mixed terms arise as part of (b) and combine to give

$$X_2 = X_1 \frac{4a^2}{3\pi} \int_0^\infty dx \left(\frac{1}{x+a} - \frac{a}{(x+a)^2} \right) \left(\sin x + \frac{3 \cos x}{x} - \frac{3 \sin x}{x^2} \right). \quad (29)$$

and this expression is C-P's $\Delta_3 E$ (given in their eqn. (40) in different form).

Since X_1 and X_2 are identical with the corresponding terms of C-P there will be no further discussion concerning them. It is in terms arising from the pure retarded field interaction that differences arise. These will be considered in greater detail.

Term (a) can derive from a variety of schemes of virtual states; one such scheme is

	Unperturbed state	Virtual states			Unperturbed state
A	0	r	0	0	0
B	0	0	0	s	0
F	0	$\mathbf{k}, \lambda$	$\mathbf{k}, \lambda + \mathbf{k}', \lambda'$	$\mathbf{k}, \lambda$	0

and the corresponding contribution to ΔE is

$$\begin{aligned} & \frac{\Sigma \Sigma \Sigma \Sigma}{r \ s \ \mathbf{k}, \lambda \ \mathbf{k}', \lambda'} \frac{-1}{\hbar^3 c^3 (k_r + k)(k + k')(k_s + k)} \times i c C_k k_r \exp(i \mathbf{k} \cdot \mathbf{R}_A)(\mathbf{q})_{0r} \cdot \mathbf{e}(\mathbf{k}, \lambda) \\ & \times -i c C_k k_r \exp(i \mathbf{k}' \cdot \mathbf{R}_A)(\mathbf{q})_{r0} \cdot \mathbf{e}(\mathbf{k}', \lambda') \\ & \times i c C_k k_s \exp(-i \mathbf{k}' \cdot \mathbf{R}_B)(\mathbf{q})_{0s} \cdot \mathbf{e}(\mathbf{k}', \lambda') \\ & \times -i c C_k k_s \exp(-i \mathbf{k} \cdot \mathbf{R}_B)(\mathbf{q})_{s0} \cdot \mathbf{e}(\mathbf{k}, \lambda). \end{aligned} \quad (30)$$

From the method of choosing the atomic wave functions this

$$= -\frac{ck_r^2 k_s^2}{\hbar^3} \sum_{i,j} \sum_{\mathbf{k},\lambda} \sum_{\mathbf{k}',\lambda'} \frac{C_k^2 C_{k'}^2}{(k_r+k)(k_s+k)(k+k')} |(\mathbf{q})_{0r}|^2 e_i(\mathbf{k},\lambda) e_i(\mathbf{k}',\lambda') \\ \times |(\mathbf{q})_{0s}|^2 e_j(\mathbf{k},\lambda) e_j(\mathbf{k}',\lambda') \exp[-i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{R}] \quad . \quad . \quad . \quad (31)$$

and replacing the summations by integrations over the spaces of $\mathbf{k}$ and $\mathbf{k}'$

$$= -\frac{k_r^2 k_s^2}{\pi^2 \hbar c R^2} |(\mathbf{q})_{0r}|^2 |(\mathbf{q})_{0s}|^2 \sum_i \int_0^\infty \frac{F dk}{(k+k_r)(k+k_s)} \int_0^\infty \frac{F' dk'}{(k+k')} \quad . \quad . \quad (32)$$

where
$$F = \left(\sin kR + \frac{\cos kR}{kR} - \frac{\sin kR}{k^2 R^2} \right)$$

when i represents the x or y direction and

$$2 \left(\frac{\cos kR}{kR} - \frac{\sin kR}{k^2 R^2} \right)$$

when it refers to the z direction.

Written in terms of the dimensionless quantities x, x' and a and allowing for the identity of the atoms the expression becomes

$$= -\frac{2a^5}{\pi^2 \hbar c a R^5} |(\mathbf{q})_{0r}|^4 \int \int \frac{MM' + 2NN'}{(x+a)^2(x+x')} dx dx' \quad . \quad . \quad (33)$$

where
$$M = \left(\sin x + \frac{\cos x}{x} - \frac{\sin x}{x^2} \right), \quad N = \left(\frac{\cos x}{x} - \frac{\sin x}{x^2} \right).$$

An identical contribution comes from a similar scheme with A and B interchanged. The two may be combined and expressed in the form

$$X_1 \frac{4a^5}{3\pi^2} \int \int \frac{MM' + 2NN'}{(x+a)^2(x+x')} dx dx' \quad . \quad . \quad . \quad (34)$$

In a similar way term (a) also gives

$$X_1 \frac{4a^5}{3\pi^2} \int \int \frac{(MM' + 2NN') dx dx'}{(x+a)(x'+a)(x+x')} \\ \text{from the scheme} \quad \begin{bmatrix} 0 & r & 0 & 0 & 0 \\ 0 & 0 & 0 & s & 0 \\ 0 & \mathbf{k} & \mathbf{k}+\mathbf{k}' & \mathbf{k}' & 0 \end{bmatrix}, \quad . \quad . \quad . \quad (35)$$

$$X_1 \frac{4a^4}{3\pi^2} \int \int \frac{(MM' + 2NN') dx dx'}{(x+a)(x'+a)}$$

from
$$\begin{bmatrix} 0 & r & r & 0 & 0 \\ 0 & 0 & s & s & 0 \\ 0 & \mathbf{k} & 0 & \mathbf{k}' & 0 \end{bmatrix} \quad \text{and} \quad \begin{bmatrix} 0 & r & r & r & 0 \\ 0 & 0 & s & 0 & 0 \\ 0 & \mathbf{k} & 0 & \mathbf{k}' & 0 \end{bmatrix}, \quad . \quad (36)$$

$$X_1 \frac{4a^5}{3\pi^2} \iint \frac{(MM' + 2NN') dx dx'}{(x+a)(x'+a)^2}$$

from $\begin{bmatrix} 0 & r & r & 0 & 0 \\ 0 & 0 & s & s & 0 \\ 0 & \mathbf{k} & \mathbf{k} + \mathbf{k}' & \mathbf{k} & 0 \end{bmatrix}$ and $\begin{bmatrix} 0 & r & r & r & 0 \\ 0 & 0 & s & 0 & 0 \\ 0 & \mathbf{k} & \mathbf{k} + \mathbf{k}' & \mathbf{k}' & 0 \end{bmatrix}$, (37)

and $X_1 \frac{8a^5}{3\pi^2} \iint \frac{xx' dx dx'}{(x+a)^2(x'+a)}$

from $\begin{bmatrix} 0 & r & r & 0 & 0 \\ 0 & 0 & s & s & 0 \\ 0 & \mathbf{k} & \mathbf{k} + \mathbf{k}' & \mathbf{k}' & 0 \end{bmatrix}$ and $\begin{bmatrix} 0 & r & r & r & 0 \\ 0 & 0 & s & 0 & 0 \\ 0 & \mathbf{k} & \mathbf{k} + \mathbf{k}' & \mathbf{k} & 0 \end{bmatrix}$, (38)

term (b) gives $-X_1 \frac{16a^4}{3\pi^2} \iint \frac{(MM' + 2NN') dx dx'}{(x+x')(x+a)}$

from $\begin{bmatrix} 0 & 0 & r & 0 \\ 0 & 0 & 0 & 0 \\ 0 & \mathbf{k} + \mathbf{k}' & \mathbf{k} & 0 \end{bmatrix}$, ... (39)

$-X_1 \frac{8a^4}{3\pi^2} \iint \frac{(MM' + 2NN') dx dx'}{(x+a)(x'+a)}$

from $\begin{bmatrix} 0 & r & r & 0 \\ 0 & 0 & 0 & 0 \\ 0 & \mathbf{k} & \mathbf{k}' & 0 \end{bmatrix}$, ... (40)

$X_1 \frac{8a^3}{3\pi^2} \iint \frac{xx' dx dx'}{(x+a)^2}$

from $\begin{bmatrix} 0 & r & r & 0 \\ 0 & 0 & 0 & 0 \\ 0 & \mathbf{k} & \mathbf{k} & 0 \end{bmatrix}$, ... (41)

and (c) gives $X_1 \frac{16a^3}{3\pi^2} \iint \frac{(MM' + 2NN') dx dx'}{(x+x')}$

from $\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & \mathbf{k} + \mathbf{k}' & 0 \end{bmatrix}$ (42)

Term (d) contains two contributions which cancel exactly with (38) and (41). Combining the remaining expressions the net contribution due to the pure retarded field interaction amounts to

$$X_3 = X_1 \frac{4a^3}{3\pi^2} \iint (MM' + 2NN') \left\{ \frac{4x}{(x+a)(x+x')} - \frac{a}{(x+a)(x'+a)} \right. \\ \left. + \frac{a^2}{(x+a)(x'+a)(x+x')} + \frac{a^2}{(x+a)^2(x+x')} + \frac{a^2}{(x+a)(x'+a)^2} \right\} dx dx', \quad (43)$$

for large values of a

$$X_3 \sim X_1 \frac{8a^3}{3\pi^2} \iint (MM' + 2NN') \frac{dx dx'}{(x+x')} \simeq 0.22a^3 X_1. \quad (44)$$

$$\text{Since} \quad X_2 \sim -2X_1 \quad \text{and} \quad \Delta E = X_1 + X_2 + X_3 \quad (45)$$

this implies that (again for large a)

$$\Delta E \sim 0.22a^3 X_1 = -\frac{0.66k_r^2}{\hbar c R^3} |(\mathbf{q})_{0r}|^4, \quad (46)$$

$$\text{i.e. that} \quad \Delta E \propto \left(-\frac{1}{R^3}\right) \text{ for large } R. \quad (47)$$

(In this connection 'large R ' means greater than a few wavelengths of the radiation emitted by the atom in making a transition from the excited to the ground state. This will normally be of the order of 10^{-5} cm.)

It will be noted that there are no infinities in the final result. Their disappearance is a consequence of ignoring contributions from the separate (A—F) and (B—F) interactions. ('P neglected the infinities on the grounds that they are independent of R ; this is legitimate reasoning but the present procedure brings out the more fundamental aspect that there are none of them specific to the interaction between the atoms.

The present result is to be contrasted with the conclusion of 'P who found that ΔE varies as $(-1/R^7)$ at large R . It should be emphasized, however, that in each case the short range approximation agrees exactly with the Eisenschitz–London result.

An obvious extension of the problem is to determine the interaction energy of two atoms moving with constant momentum. This is under consideration and it is hoped to publish the results in a further communication.

ACKNOWLEDGMENTS

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CXLIV. *K-capture in the Decay of Chlorine 36*

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ABSTRACT

A search for the existence of K-capture in ^{36}Cl has been made using a proportional tube spectrometer with labelled methyl chloride as a gaseous source. A peak which was observed in the beta spectrum corresponded to the K-radiation of sulphur and indicated the presence of K-capture. The branching ratio K/β^- was found to be $(1.7 \pm 0.1)\%$. A scintillation spectrometer failed to detect any gamma-ray lines above 20 kev with intensity greater than 10^{-4} of the beta intensity, so the observed K-capture transition goes to the ground state of ^{36}S .

§ 1. INTRODUCTION

THE isotope ^{36}Cl is known to be an electron emitter with a half-life of $(3.08 \pm 0.03) \times 10^5$ years (Bartholomew, Boyd, Brown, Hawkings, Lounsbury and Merritt 1955) and with a maximum energy of (714 ± 5) kev (Feldman and Wu 1952). No gamma-rays with intensity greater than 5% of the beta particles are present (Wu, Townes and Feldman 1949). The spins of the initial and final nuclei are 2 and 0 respectively (Townes and Aamodt 1949) and the ft-value is 3×10^{13} sec. The transition is second forbidden and the spectrum shape has been extensively studied by Wu and co-workers (Longmire, Wu and Townes 1949), as it allows a detailed analysis of the kind of interaction (see Wu 1955). The observed data can be fitted on the basis of a no parity change transition and a tensor interaction.

^{36}Cl is the lightest nucleus which has two neighbouring stable isobars, and it might decay both to ^{36}A by electron emission and to ^{36}S by positron emission or electron capture. However, no positrons have been observed in experiments which could have detected 10^{-4} positron per beta disintegration (Wu, Townes and Feldman 1949). Recent mass measurements (Wapstra 1955) give 380 kev for the energy available in the transition to ^{36}S . Electron capture is, therefore, possible and a search for this was made in the present work.

§ 2. EXPERIMENTAL PROCEDURE

Since there are no gamma rays, K-capture can only be detected by direct measurement of the sulphur K x-rays, which have an energy

* Communicated by Professor P. I. Dee, F.R.S.

of 2.3 kev. For this purpose the proportional tube is the most suitable type of spectrometer. However, a solid source would not give reliable results since 95% of the K-capture transitions must result in emission of Auger electrons and large errors due to scattering and absorption would occur. By using a gaseous source in the proportional counter (see Curran 1955) these errors are eliminated, and in the present experiments this technique was employed.

Chlorine itself or hydrogen chloride could not be used as gaseous sources since they reacted with the counter walls. However, methyl chloride was found to be very suitable. On opening a counter containing this source to a vessel of equal volume, the activity was exactly halved, indicating that none of the source was on the walls.

The chlorine 36 was supplied by Atomic Energy of Canada Limited, as a processed isotope of specific activity $120 \mu\text{C/g}$ of chlorine in the form of hydrochloric acid free from radioactive impurities. Chlorine liberated from the acid was used to prepare methyl chloride by photochlorination of methane. Hydrogen chloride and chlorine were removed by reaction with sodium bicarbonate and mercury.

A brass counter of diameter $d=10$ cm and of sensitive length $l=58$ cm, fitted with field correcting tubes, was employed. It was filled with an argon-methane mixture to pressures up to 2 atmospheres. A suitable amount of methyl chloride was introduced by diffusion after filling was complete.

The electronic equipment included a Hutchinson-Scarrott multi-channel kicksorter. The 9.2 kev x-rays from an external ^{71}Ge source were used for energy calibration. In addition the resolution of the x-ray line provided a convenient check of the operation of the counter. No change in the shape of the calibration peak was observed when the methyl chloride was introduced.

Since the beta particles from the chlorine produced very large pulses, counting rates were kept below 100 counts/sec, and the cosmic-ray background was reduced by extensive lead shielding.

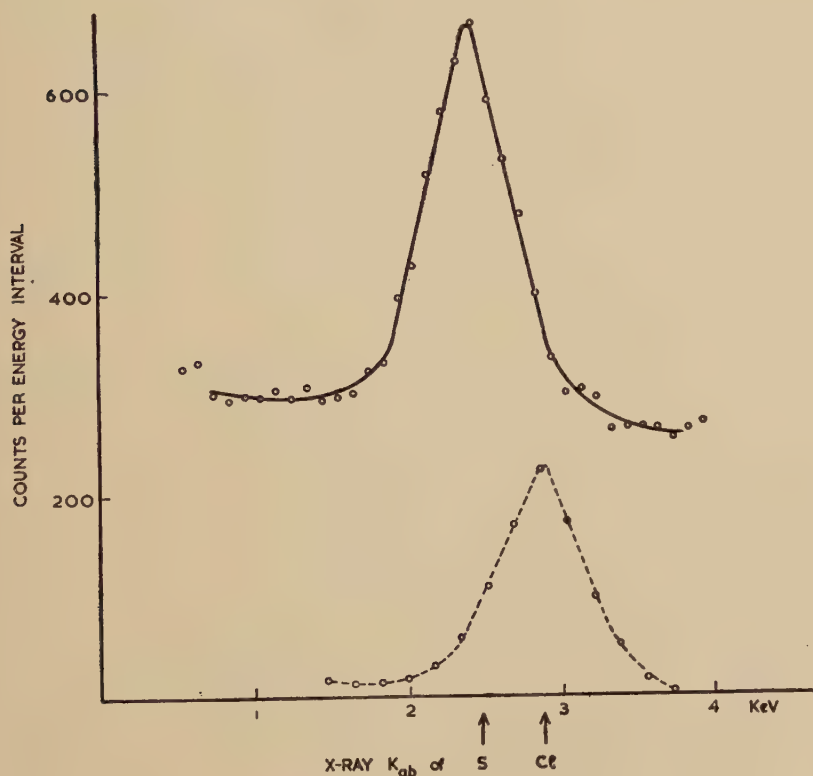
As no change of counting rate was detected in a period of 2 weeks the absence of radioactive impurities was established. The possible impurities are of relatively short half-life and they would have been revealed by a decay of counting rate.

Part of a typical spectrum, in the range between 0.5 kev and 4 kev is shown in fig. 1. A peak at about 2.4 kev, superimposed on a continuous spectrum due to the beta particles, is evident. In order to obtain a more accurate energy calibration in this region a trace of ^{37}A was allowed to diffuse into the counter after the spectrum measurements had been completed. This source decays by K-capture and emits the 2.8 kev radiation of chlorine. The ^{37}A calibration peak obtained is shown dotted in fig. 1, the scale on vertical axis being reduced by a factor of 100. This shows that the peak from the ^{36}Cl source had exactly the same energy as sulphur K-radiation. The ratio of the number of counts in the peak

to the total number of counts from the source was 1.65%. In determining the area of the ^{36}Cl peak the base of the peak was established by comparison with the ^{37}A calibration spectrum.

The presence of the sulphur x-ray peak is definite evidence for K-capture of ^{36}Cl since in the above experiment such a peak cannot be produced

Fig. 1



Part of the spectrum of ^{36}Cl showing the peak due to sulphur x-rays. The number of pulses in the whole spectrum is 182 000. The dotted curve is a calibration peak from an ^{37}A source.

in any other way. It is worth pointing out that x-rays from excited atoms produced in the ^{36}Cl beta decay have an energy equal to that of argon K x-rays, and are not detected with a gaseous source since they are effectively simultaneous with the beta particles.

§ 3. BRANCHING RATIO

In order to obtain the branching ratio between K-capture and beta emission the value given above for the relative number of counts in the peak compared with the number of counts in the total spectrum must be corrected for errors introduced by the finite dimensions of the counter,

The number of counts in the peak is affected since some of the x-ray photons escape from the sensitive volume of the counter, while some of those produced outside cause photoelectrons within it and are detected. Further, the observed number of beta disintegrations is too large on account of electrons entering the sensitive volume of the counter from the ends of the counter. Both these corrections can be calculated rigorously, but in the present experiments they are relatively small and it is sufficient to estimate them approximately.

Due to the Auger effect x-ray photons are emitted in only 5.6% of the K-capture disintegrations. For the sulphur K x-rays the half-thickness in argon at 2 atmospheres is 0.54 cm. Since the distance between the boundaries of the sensitive volume and the ends of the counter is $h=6.5$ cm which is large compared with the half-thickness, photons entering the counting volume compensate for those leaving it. The probability of escape of photons through the side walls of the counter has been estimated to be 5%. The total correction to the K-capture due to the photons is, therefore, 0.3%.

In the case of the beta particles the error in counting rate is due to that part of the source which is outside the sensitive volume of the counter, and is evidently less than h/l , which in the counter used is 11%. In a first approximation for the calculation of the error the absorption of electrons can be neglected since h is appreciably smaller than the average range of the electrons, and effective solid angles can be approximated for different elements of volume and integrated. The values so obtained can be corrected for the absorption of the electrons on the basis of the known beta spectrum. Such a calculation gave a value of 4.5% for the beta intensity correction.

After applying both corrections and taking a weighted mean of several measurements, the branching ratio $K\beta$ was found to be $(1.7 \pm 0.1)\%$.

It should be remarked that no measurement of L-capture, which will increase the total branching ratio slightly, has yet been made.

§ 4. DECAY SCHEME

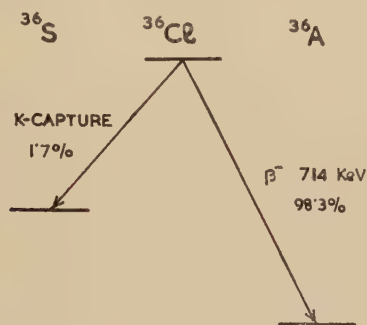
In order to establish the decay scheme a search for gamma-rays that might be associated with the K-capture was made using a scintillation spectrometer. A ^{36}Cl source of $1\mu\text{C}$ activity in the form of hydrogen chloride, surrounded by a polythene beta absorber, was mounted in front of a NaI(Tl) crystal of length and diameter equal to 5 cm. No gamma-ray peaks were observed above the continuous bremsstrahlung spectrum. Since any gamma peak in the energy region above 20 kev with an intensity greater than 1% of the total bremsstrahlung would have been detected, the experiment sets an upper limit for gamma-ray intensity of 10^{-4} gamma per beta particles.

However, any low energy gamma rays present would probably be strongly internally converted. The energy region below 50 kev was, therefore, investigated with the proportional counter containing the

gaseous source. Apart from the K-capture peak no other peaks were observed above the beta spectrum. However, since the spectrum is completely distorted and is recorded mainly in the region below 100 kev (as most of the beta particles spend only a small fraction of their energy in the sensitive volume) peaks with energy above 15 kev of intensity less than 1% of the total counting rate could not have been distinguished from the large background. Even if any such converted gamma-rays are present the K-capture associated with them would not be recorded in the peak shown in fig. 1 because the counter would give a pulse corresponding to the sum of both energies.

It may be concluded that the observed electron capture transition which occurs in 1.7% of all disintegrations goes to the ground state of ^{36}S and the decay scheme is as shown in fig. 2.

Fig. 2

Decay scheme of ^{36}Cl .

§ 5. REMARKS ON THE USE OF A SOLID SOURCE

Some results obtained with a very thin solid source of ^{36}Cl prepared by reaction of chlorine gas with the walls of a soft glass counter of external cathode type are worth mentioning. As $30\ \mu\text{g}$ of chlorine were deposited on a surface of $1000\ \text{cm}^2$ the average source thickness is $0.03\ \mu\text{g}/\text{cm}^2$ if uniform spreading is assumed. The observed K-capture peak had an intensity of less than 10% of that obtained with a gaseous source in the same counter and was shifted to a lower energy. The result shows that even very thin solid sources deposited on the wall are not reliable in this very low energy region. Further work on this phenomenon will be discussed separately.

ACKNOWLEDGMENTS

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CXLV. *Surface Distributions of Dislocations in Metals*

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[Received August 10, 1955]

ABSTRACT

It is shown that a boundary containing dislocations of two systems can, in general, form on two, and only two, planes, which have simple indices, and that the rotation axes are simple crystallographic directions. Most of the experimental evidence on misorientations in deformed crystals can be explained in terms of such simple boundaries.

§ 1. INTRODUCTION

RECENT work (Hirsch and Kellar 1952, Gay and Kelly 1953 a, b) has shown that many of the dislocations left inside a metal after deformation are arranged along surfaces forming low angle boundaries between neighbouring regions of crystal, these latter being called subgrains, cells or particles. Such low angle boundaries are observed after either high temperature or room temperature deformation. The effect of these particles on the mechanical properties is not clear, although various suggestions have been made (Bragg 1949, Mott 1952, Gay, Hirsch and Kelly 1954). To understand their effect, the nature and method of formation of the boundaries must be known. While the size of the particles is generally well established, little is known about the misorientations and the arrangement of the dislocations in the boundaries. In the course of the deformation of single or polycrystals, slip may take place on one or more slip systems in the same region of crystal. It is likely that the dislocations will tend to form surface arrays in order to reduce the strain energy. In this paper an investigation is made of the types of stable boundary that can be formed with dislocations of one or two systems: certain special cases involving more than two sets of dislocations will also be considered. The experimental data are discussed in the light of the theoretical predictions. It is hoped in a later paper to consider the case of slip on three or more systems.

§ 2. THEORY

Frank (1950) has shown that the dislocation content of a plane low angle boundary can be obtained from the equation

$$\mathbf{d} = (\mathbf{r} \times \mathbf{l}) 2 \sin \frac{1}{2} \alpha \quad . \quad . \quad . \quad . \quad . \quad (1)$$

* Communicated by the Authors.

where $\mathbf{d}$ is the vector sum of the Burgers vectors of the dislocations intersected by a line $\mathbf{r}$ lying in the boundary, $\mathbf{l}$ is a unit vector parallel to the axis of rotation and α is the angular rotation about this axis. It follows from (1) that $\mathbf{d}$ is normal to $\mathbf{l}$ and $\mathbf{r}$ for all $\mathbf{r}$. Further the dislocation line must lie in the boundary plane and in the slip plane which is assumed to be one of the commonly observed slip planes.* These conditions define the boundary plane and the axis of rotation of a boundary formed with dislocations of one or two systems. The actual dislocation content may then be found easily by application of eqn. (1).†

Case 1. Slip on One System

Since $\mathbf{r}$ is normal to $\mathbf{d}$ for all $\mathbf{r}$, and $\mathbf{d}$ is fixed in direction, the boundary plane must be normal to this direction, i.e. the direction of the Burgers vector, and so is normal to the slip plane. The dislocation lines are all parallel to the intersection of the slip and boundary planes: if we take $\mathbf{r}$ parallel to the dislocation lines, $\mathbf{d}=0$, and so $\mathbf{l}$ must also be parallel to this direction.

This boundary is a pure tilt boundary.

Case 2. Slip on Two or More Systems in One Plane

As in the previous case, if the boundary is not parallel to the slip plane, it must be normal to it. $\mathbf{l}$ is then again parallel to the dislocation lines, and the orientation of the boundary is determined by the relative numbers of dislocations of the different types, which can vary. The boundary is again a pure tilt boundary.

If the boundary is parallel to the slip plane, since $\mathbf{r}$ and $\mathbf{d}$ are both always in the slip plane, $\mathbf{l}$ is normal to this plane. The boundary is a pure twist boundary.

Case 3. Slip on Two or More Systems with One Burgers Vector

$\mathbf{r}$ is normal to $\mathbf{d}$ and, as in case 1, $\mathbf{d}$ is fixed in direction, so the boundary plane must again be normal to the Burgers vector of the dislocations. The boundary is a pure tilt boundary, but the axis of rotation may take up any position within this plane, depending on the relative contributions to the boundary of the dislocations on the various slip planes.

Case 4. Slip on Two Unrelated Systems

(a) If the dislocations are not all parallel it is convenient to choose $\mathbf{r}_1$ parallel to one set of dislocations, so that it cuts only the other set of dislocations. $\mathbf{d}_1$ is then parallel to $\mathbf{b}_2$, the Burgers vector of the second

* For boundaries formed under conditions where an unlimited amount of climb is permitted, see Frank (1955).

† Note that for a given pair of systems, the ratio of the numbers of dislocations of each system is fixed.

set. Then it follows from (1) that $\mathbf{l}$ is normal to both Burgers vectors. The direction of the dislocation lines is defined since $\mathbf{r}_1$ must be normal to $\mathbf{b}_2$: the boundary contains both sets of dislocation lines, and therefore it also is uniquely determined.

(b) If the dislocations are parallel to each other, they must be parallel to the intersection of the slip planes and it follows that $\mathbf{l}$ is also parallel to this direction. Since $\mathbf{l}$ lies in the boundary, $\mathbf{d}$ must be perpendicular to the boundary. Suppose that the boundary contains n_1, n_2 dislocations per cm normal to the dislocation lines with Burgers vectors $\mathbf{b}_1, \mathbf{b}_2$ respectively. Then $(n_1\mathbf{b}_1 + n_2\mathbf{b}_2)$ must be normal to $\mathbf{l}$, so $n_1\mathbf{b}_1 \cdot \mathbf{l} = -n_2\mathbf{b}_2 \cdot \mathbf{l}$. Thus the relative dislocation content of the boundary is determined, and, since $(n_1\mathbf{b}_1 + n_2\mathbf{b}_2)$ is normal to the boundary, the orientation of the boundary is uniquely defined.

Case 5. Slip on Several Planes with coplanar Burgers Vectors

For all positions of $\mathbf{r}$ in the boundary, $\mathbf{d}$ is in the plane containing the Burgers vectors; hence $\mathbf{l}$ is normal to this plane. No simple generalization seems possible about the orientation of the boundary plane.

If the slip planes lie in a zone, a special case may arise in which the dislocations are all parallel to each other and to the zone axis. The axis of rotation is also parallel to this direction. The boundary is a pure tilt boundary.

§ 3. APPLICATIONS

3.1. Hexagonal Systems

At room temperature the operative slip systems in hexagonal metals are (0001) [1210], while at high temperatures slip can apparently also take place on (1010), (1011) [1210] (Boas and Schmid 1930, Kolesnikov 1938, Cahn, Bear and Bell 1954). The stable boundaries for various combinations of slip systems are given in table 1. The boundary of case 1 corresponds to the frequently observed 'bend plane' (Hess and Barrett 1949, Orwan 1942).

Cahn, Bear and Bell (1954) report the presence of pure tilt boundaries of arbitrary orientation normal to the basal plane in polycrystals of Zn deformed by creep at 250°C, the axis of rotation lying in the basal plane (fig. 1). This corresponds to case 2. It should be noted that although the boundaries are approximately pure tilt boundaries, in general each boundary consists of at least two sets of mixed type dislocation with different Burgers vectors. (The description given by the authors is incorrect.) It is interesting to observe, however, that the mean orientation of the rotation axis appears to be about 5° off the expected (0001) plane. For polycrystalline Zn deformed by creep at 350°C, Cahn, Bear and Bell report the presence of tilt boundaries with rotation axes shown in fig. 2, and the occurrence of slip on (1010) and (1011) planes. The expected positions for the rotation axes are also marked on fig. 2. Although a

number of points lie close to the expected positions, it is clear that some of the results do not conform with the theory.

Table 1. Rotation Axes in Hexagonal Metals

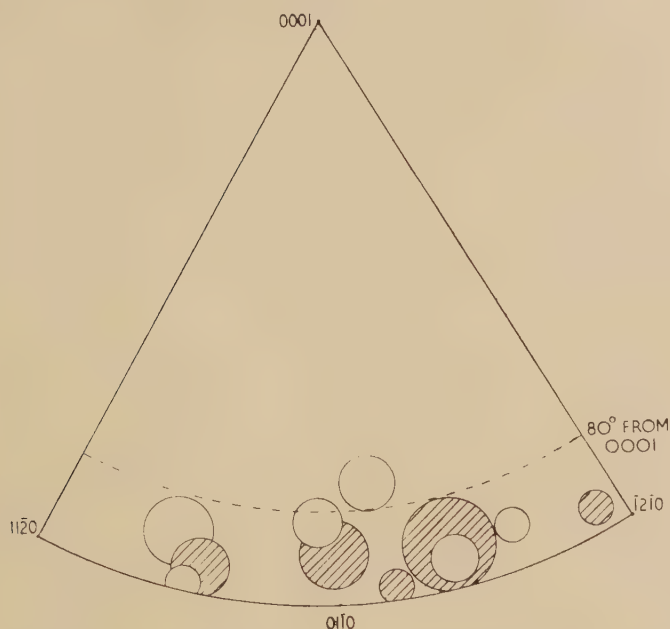
	Slip plane	Slip direction	Rotation axis	Boundary plane	Type of boundary
Case 1	(0001)	$[\bar{1}210]$	$[10\bar{1}0]$	$(10\bar{1}0)$	tilt
Case 2	(0001)	several $[\bar{1}210]$	(a) $[hk.o]$ (b) $[0001]$	$[\bar{k}h.o]$ $[0001]$	tilt twist
Case 3	(0001) (10 $\bar{1}0$)	$[\bar{1}210]$ $[\bar{1}210]$	$[10\bar{1}l]$	$(\bar{1}210)$	tilt
	(0001) (10 $\bar{1}1$)	$[\bar{1}210]$ $[\bar{1}210]$	$[10\bar{1}l]$	$(\bar{1}210)$	tilt
Case 4	(0001) (1 $\bar{1}00$)	$[\bar{1}210]$ $[\bar{1}120]$	$[0001]$	$(11\bar{2}0)$	tilt
	(0001) (10 $\bar{1}0$)	several $[\bar{1}210]$ $[\bar{1}210]$	$[\bar{1}210]$	$(10\bar{1}0)$	tilt
	(0001) (10 $\bar{1}1$)	several $[\bar{1}210]$ $[\bar{1}210]$	$[\bar{1}210]$	$(10\bar{1}0)$	tilt
	A (10 $\bar{1}0$) (01 $\bar{1}1$)	$[\bar{1}210]$ $[\bar{2}1\bar{1}0]$	$[01.\bar{1}]$	$(10\bar{1}0)$	tilt
	B (10 $\bar{1}1$) (01 $\bar{1}1$)	$[\bar{1}210]$ $[\bar{2}1\bar{1}0]$	$[11.\bar{1}]$	$(1\bar{1}00)$	tilt
	C (10 $\bar{1}1$) (1101)	$[\bar{1}210]$ $[\bar{1}120]$	$[12.\bar{1}]$	$(2\bar{1}\bar{1}0)$	tilt
Case 5	one or several (0001) (10 $\bar{1}0$) (10 $\bar{1}1$)	several $[\bar{1}210]$	$[0001]$	depends on dislocation content	dislocation

Note.—A, B, C are marked on fig. 2.

The discrepancies between theory and experiment may be explained in the following way. It has been assumed that the dislocation lines are straight and that they lie completely in their usual slip plane. It is possible, however, that the mean direction of a dislocation line may not lie in any simple crystallographic plane if it contains a large number of jogs.* Figure 3 shows a possible arbitrary configuration of a dislocation line.

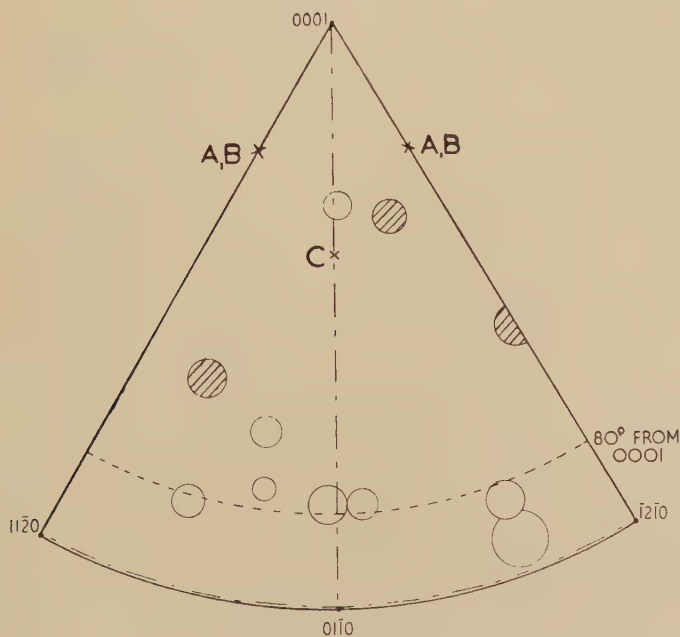
* The jogs might be formed by cross slip or climb.

Fig. 1



Tilt axes in Zn after creep at 250°C (Cahn, Bear and Bell 1954). Shaded circles refer to axes deduced from photographs with only two sub-spots to each Laue spot.

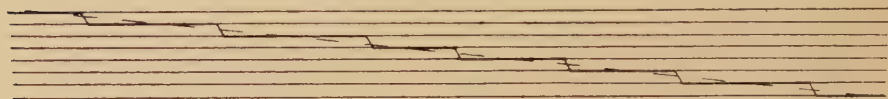
Fig. 2



Tilt axes in Zn after creep at 350°C (Cahn, Bear and Bell 1954). Rotation axes predicted by theory are indicated thus x, ————. Letters refer to corresponding case in table 1. Shaded circles refer to axes deduced from photographs with only two sub-spots to each Laue spot.

A boundary formed with parallel dislocations of this kind, with the same or different Burgers vectors, would have an axis lying at an angle of, say, 10° to the ideal orientation. Larger numbers of jogs or possibly slip on planes such as $(10\bar{1}2)$ or $(10\bar{1}3)$ could account for the larger deviations from the simple theory at the higher temperature.

Fig. 3



Possible configuration of dislocation line containing a large number of jogs.

3.2. Face-Centred Cubic System

At room temperature the slip systems are $\{111\} [\bar{1}10]$. Table 2 gives all the possible boundary configurations for slip on one or two systems. Apart from the well known kink bands of case 1 (Cahn 1951, Honeycombe 1950) there is little experimental evidence on misorientations in deformed

Table 2. Rotation Axes in Face-Centred Cubic Metals

	Slip plane	Slip direction	Rotation axis	Boundary plane	Type of boundary
Case 1	(111)	$[\bar{1}10]$	$[11\bar{2}]$	$(\bar{1}10)$	tilt
Case 2	(111)	several $[\bar{1}10]$	(a) in (111) (b) $[\bar{1}11]$	$\perp(111)$ (111)	tilt twist
Case 3	<i>d</i> (111) ($\bar{1}11$)	$[0\bar{1}1]$ $[0\bar{1}1]$	in $(0\bar{1}1)$	$(0\bar{1}1)$	tilt
Case 4	<i>e</i> (111) ($\bar{1}11$)	$[10\bar{1}]$ $[0\bar{1}1]$	(a) $[\bar{1}11]$ (b) $[0\bar{1}1]$	$(13\bar{1})$ (211)	mixed tilt
	<i>f</i> (111) ($\bar{1}11$)	$[101]$ $[101]$	(a) $[010]$ (b) $[0\bar{1}1]$	(010) (100)	twist tilt
	<i>g</i> (111) ($\bar{1}11$)	$[101]$ $[\bar{1}10]$	(a) $[\bar{1}11]$ (b) $[0\bar{1}1]$	(111) (011)	mixed tilt
Case 5	several (111)	coplanar $[\bar{1}10]$	$[\bar{1}11]$	—	mixed in general

Notes.—*d*, primary and cross slip.

f, primary and conjugate slip in compression.

g, primary and conjugate slip in tension.

g(b), dislocations can react to form Cottrell-Lomer locks.

crystals. Barrett and Levenson (1940) report the presence of deformation bands parallel to (010) for compressed Al crystals. The primary and conjugate systems for this type of deformation would result in just this type of boundary. Ball (unpublished work) has recently examined with a microbeam a polycrystal of Al extended at 300°C. The Laue photograph from a region showing slip on two sets of planes consisted of spotty streaks indicating a unique axis of misorientation which was found to be very near to [100]. This again corresponds to the type *f* boundary of table 2. Evans, Layton and Wilman (1951) have reported the existence of the axes of rotation shown in table 3 in abraded crystals of copper. The boundary

Table 3

Crystal surface	Direction of abrasion	Axis of rotation
(110)	[1 $\bar{1}$ 0]	[001]
(110)	[001]	[1 $\bar{1}$ 0]
(110)	[112]	[1 $\bar{1}$ 1]

planes were not determined. Wilman has postulated a new slip mechanism, 'rotational slip', to account for these axes of rotation (Evans, Layton and Wilman 1951). It is clear, however, that all these axes of rotation are expected to occur if slip takes place on two of the usual translational slip systems. In particular, the rotation about [1 $\bar{1}$ 1] could be obtained with a number of different pairs of slip systems, contrary to the statement made by Evans, Layton and Wilman. It should be noted that although the [100] and [1 $\bar{1}$ 1] rotations could correspond to twist boundaries, the rotation about [1 $\bar{1}$ 0] occurs at a tilt boundary (see table 2). A determination of the boundary planes would therefore serve as a test of the 'rotational slip' hypothesis and of the explanation suggested here.

Wilman (1951) also reports rotation about [001] in a number of beaten and annealed foils of face-centred cubic metals (Cu, Ag, Au, Al, Pd, Pt). He observes that in many cases the boundary angles are such that there is a simple periodic fit between atoms on the two sides of the boundary, assuming this to be (001). The theory presented in this paper predicts (001) as the boundary plane, so Wilman's observations may be considered as evidence for the existence of cusps in the curve of energy versus boundary angle, as predicted by dislocation theory (e.g. Read 1953).

3.3. Body-Centred Cubic System

At room temperature the operative slip systems appear to be (110), (11 $\bar{2}$), (123) [111], and of these (1 $\bar{1}$ 0) [111] is probably the most important. Table 4 gives the type of stable boundary for slip on (110) [111] systems, and a special case for the other systems.

Deformation bands with planes parallel to (111) have been observed for Fe and β brass. There is very little evidence on boundaries containing

more than one set of dislocations. Barrett and Levenson (1939) report the occurrence of deformation bands parallel to (100) and (111) for crystals of Fe deformed in tension (cases *k*, *h*). Evans, Layton and Wilman (1951)

Table 4. Rotation Axes in Body-Centred Metals

	Slip plane	Slip direction	Rotation axis	Boundary plane	Type of boundary
Case 1	(110) (112) (123)	[111] [111] [111]	[112] [110] [541]	(111) (111) (111)	tilt tilt tilt
Case 2	(110)	[111] and [11 $\bar{1}$]	(a) in (110) (b) [110]	1(110) (110)	tilt twist
Case 3	<i>h</i> (110) (101)	[111] [111]	in (111)	(111)	tilt
Case 4	<i>i</i> (110) (101)	[111] [111]	(a) [011] (b) [111]	(111) (011)	mixed tilt
	<i>j</i> (110) (101)	[111] [111]	(a) [110] (b) [111]	(131) (112)	mixed tilt
	<i>k</i> (110) (110)	[111] [111]	(a) [101] (b) [001]	(001) (010)	mixed tilt
Case 5	one or several (110) (112) (123)	[111] [111]	[110]	—	mixed in general

Notes.—*h*, primary and cross slip.

i, primary and conjugate slip in compression.

k, primary and conjugate slip in tension.

k(b), dislocations may react in a manner analogous to the formation of Lomer locks in the face-centred cubic system.

have observed the rotation axes given in table 5 for abraded crystals of Fe. These misorientations may again be readily explained in terms of boundaries consisting of two sets of dislocations (table 4).

Table 5

Crystal surface	Abrasion direction	Rotation axis
(110)	[110]	[001]
(110)	[112]	[111]
(001)	[110]	[110]

§ 4. DISCUSSION

It appears that many of the experimental data obtained so far on the nature of dislocation boundaries in deformed crystals can be explained on the assumption that slip takes place on one or two of the well-established slip systems, and that Frank's formula is applicable. This latter formula, which is used as the criterion for stability of the dislocation boundaries in this paper, gives the arrangement of dislocations necessary to fit together two unstressed regions of crystals misorientated relative to one another. It follows that the dislocations are always arranged in such a way that their long range stresses cancel out, and such a configuration has a minimum potential energy. This interpretation of the stability condition may be illustrated by the following example. Suppose in the case of a hexagonal close-packed metal slip takes place on several systems in (0001), and suppose that an arbitrary boundary is formed. This boundary has in general a resultant screw component parallel to the dislocation lines, and an edge component in the slip plane perpendicular to the dislocations. The long range stresses of an edge component can be annulled by allowing the dislocations to lie in a plane normal to (0001); this leaves a total screw component, equivalent to a pile up of screws, the long range stresses of which could only be cancelled out by slip taking place with a Burgers vector not lying in (0001). Since this is not possible the dislocation wall is orientated in such a way that the resultant screw component is zero. Frank's formula gives precisely this condition.

If there are any external stresses acting on the grains, it is possible that the dislocation boundaries may be forced out of their equilibrium position. A similar effect may occur if a cell structure is formed, for the minimum energy dislocation boundaries of such a structure may not be compatible with each other. The long range stresses of such boundaries may be eliminated by slip on further systems or climb of a suitable amount. Deviations from the simple theory can apparently be explained in this way.

With regard to the cellular structure formed during deformation at room temperature (Hirsch and Kellar 1952, Gay and Kelly 1953 a, b) there is as yet no clear evidence about the nature of the rotations; from the theoretical point of view, a cellular structure may require slip on more than two systems and the stability conditions may have to take into account the mutual compatibility of the boundaries. It is hoped to consider these points in a later paper.

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CXLVI. *The Anisotropic Absorption of the Visible Bands
in Irradiated α -Quartz*

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ABSTRACT

Measurements have been made, using polarized light, of the anisotropy of the visible absorption of centres induced in quartz by x-ray, gamma and neutron irradiation. The ratio of the integrated absorption when the electric vector is parallel to the c -axis to that when it is perpendicular, is 1.5. An experiment is described which suggests that the optical absorption is associated with substitutional aluminium impurity. It is shown that a value of 1.3 would be expected from the O'Brien and Pryce model of the magnetic resonance centre which is induced under similar conditions.

§ 1. INTRODUCTION

THE anisotropic absorption of plane polarized light by absorbing centres in solids indicates that the wave function of at least one of the states involved in the transition has directional properties related to the axes of the crystal. That such an effect occurred in the visible bands (A_1 and A_2) induced in quartz by irradiation was suggested by the work of Ditchburn *et al.* (1954). They found that the shape of the absorption curve and the values of the absorption coefficients depended on the orientation of the crystal relative to the incident beam.

The results of measurements made using plane polarized light, which are described in this paper, show that the centre is in fact anisotropic. The anisotropy accounts for the directional effect observed by Ditchburn *et al.* and is in reasonable quantitative agreement with that derived from the model which O'Brien and Pryce (1954) use to explain the paramagnetic resonance results of Griffiths *et al.* (1954).

§ 2. MEASUREMENT OF THE POLARIZATION EFFECTS

The determination of the complete polar diagram, of absorption as a function of the direction of the electric vector $\mathbf{E}$, for an induced absorbing centre in quartz is complicated by the natural birefringence and optical rotation of the crystal. However, for the major effects these factors are shown to be negligible or can be taken into account.

The types of measurement carried out are summarized in table 1. Types (i), (ii) and (v) can all be carried out on the same parallel cut

* Communicated by Professor R. W. Ditchburn.

specimen, that is cut so that the c -axis is in the plane of the polished surfaces. We have prepared two specimens each having two pairs of polished faces in order that all types of measurement could be made on the same specimen. This is particularly important when discussing the positions of bands since only then is it certain that all the other conditions are identical, in particular the specimen and the irradiation. X-ray, γ -ray and neutron irradiations were used to induce the absorption in specimens cut from three crystals of Brazilian quartz.

Table 1. Types of Measurement

Type	Direction of $\mathbf{E}$	Direction of incident light
(i) (ii)	Plane polarized, $\mathbf{E} \parallel c$ -axis Plane polarized, $\mathbf{E} \perp c$ -axis	$\perp c$ -axis
(iii) (iv)	Plane polarized, $\mathbf{E} \perp c$ -axis Unpolarized, but $\mathbf{E}$ always $\perp c$ -axis	$\parallel c$ -axis
(v)	Unpolarized, $\mathbf{E}$ at all angles to c -axis	$\perp c$ -axis

All the measurements have been made with a Hilger 'Uvispek' spectrophotometer. For the experiments involving plane polarized light a polaroid disc, which was attached to the housing of the exit slit of the monochromator, could be rotated in the beam. The polarization of the light transmitted by the polaroid exceeded 99.8% except near the extreme limits of the visible spectrum. The plane of polarization could be set to about $\pm \frac{1}{2}^\circ$.

In order to eliminate any effect associated with the rotation of the plane of polarization by the crystal and the selective response of the photocell, the latter has been determined. The polarizer was set so that $\mathbf{E}$ was vertical. Let I_0 be the intensity of plane polarized light which, for the first measurement, fell on the cell giving a response kI_0 , where k is a constant. An unirradiated quartz specimen, cut so as to give a rotation $[\alpha(\lambda)]$ of 80° at $6600 \text{ \AA}$, transmitted an intensity I_t when placed in the beam. The response of the cell was then kI_tR_α , giving a measured transmission $T_1 = T_0R_\alpha$, where $T_0 = I_t/I_0$ and R_α is the ratio of the response of the cell to plane polarized light having $\mathbf{E}$ at an angle $\alpha(\lambda)$ from the chosen initial direction. This was repeated for settings of the polarizer such that the incident $\mathbf{E}$ was $n \cdot \alpha(\lambda)$ from the vertical, where n is an integer. When $n=1$ we have

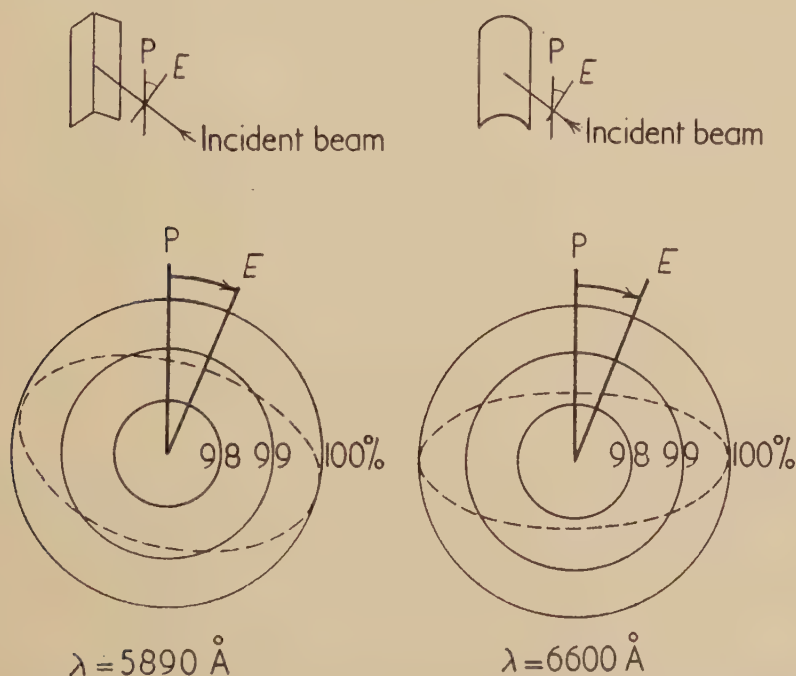
$$T_2 = \frac{I_t \cdot R_{2\alpha(\lambda)}}{I_0 \cdot R_{\alpha(\lambda)}} = \frac{T_0^2}{T_1} \cdot R_{2\alpha(\lambda)} \quad \text{or} \quad R_{2\alpha(\lambda)} = \frac{T_1 T_2}{T_0^2}$$

and for n

$$R_{(n+1)\alpha(\lambda)} = \frac{T_1 T_2 \dots T_{n+1}}{T_0^{n+1}}.$$

From measurements of T the values of $R_{\alpha(\lambda)}$ were found for the two photocells for different wavelengths and some of the results are shown in fig. 1. The value of T_0 computed from these results, when $(n+1)\alpha(\lambda)=180$, agreed within 0.5% with the value calculated from refractive index data. The geometry of the photocell surfaces is also indicated in fig. 1. It will be seen that the response was greatest when the incident $\mathbf{E}$ had a component perpendicular to the surface of the cell.

Fig. 1

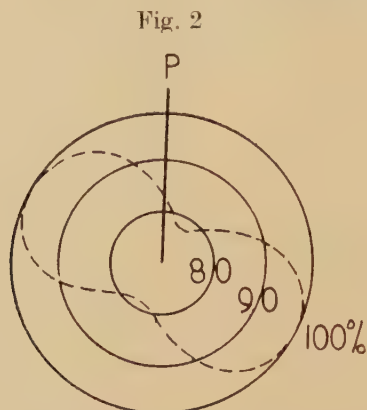


The relative response (R_{α} in %, broken line) of the photocells to plane polarized light having $\mathbf{E}$ at an angle to the vertical $\mathbf{P}$. The geometry of the emissive surfaces is shown above the polar diagrams, the L.H. cell being normally used from 2100 to 6500 $\text{\AA}$ and the R.H. cell from 6000 to 10000 $\text{\AA}$.

Various type (iv) and (v) measurements have been reported for smoky quartz, e.g. Cohen (1954), and the results are similar to the previously mentioned type (iv) and (v) measurements of Ditchburn *et al.* (1954) on irradiated quartz. To compare these results quantitatively with the type (i), (ii) and (iii) measurements the polarization introduced by the monochromator must be determined. In our case the light is partially polarized by the six reflections in the monochromator.

Measurements were made with the polaroid attachment in the beam, the response of the photocell being observed as the setting of the polaroid was varied. From the known selective response of the cell it was then possible to determine the variation of the relative intensity of radiation

resolved into a given plane of polarization, as a function of the angular variation of the plane. A typical result is shown in fig. 2. The orientation of the axis of the figure relative to the exit slit was found to depend on the wavelength, probably owing to the rotation of the plane of polarization in two crystalline quartz lenses in the instrument.



The polarization figure of the monochromator at 6100 Å. The radii scale is in % of maximum polarization.

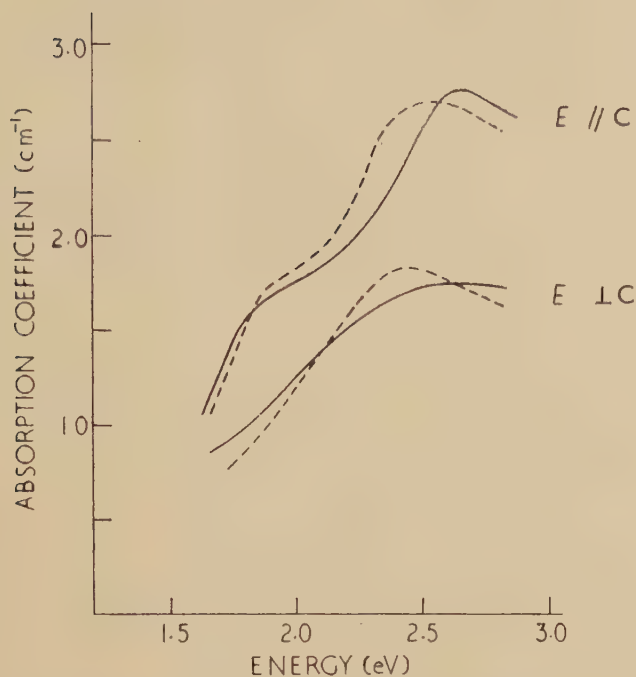
§ 3. RESULTS

The full line absorption curves in fig. 3 correspond to measurements of types (i) and (ii) in table 1. It will be seen that the absorption is greater when **E** of the incident light is parallel to the *c*-axis of the crystal than when it is perpendicular. In addition the A_1 band (2.0 eV) is more pronounced, being hardly detectable when **E** is perpendicular to the axis. At -195°C the A_2 peaks (about 2.7 eV) on both curves have moved to lower energies and the A_1 band on the higher absorption curve is less well marked.

In fig. 4 absorption curves are shown for measurements of types (iii), (iv) and (v) together with the curves (broken lines) for (i) and (ii), all referring to the same specimen. Using the results for the instrumental polarization and the selective response of the cell it was found that the instrumental effect arising because of the natural rotation was negligible for type (iv) measurements. On the other hand for (iii), in which the light was deliberately plane polarized, a small correction had to be made; the curve shown in fig. 4 has been corrected.

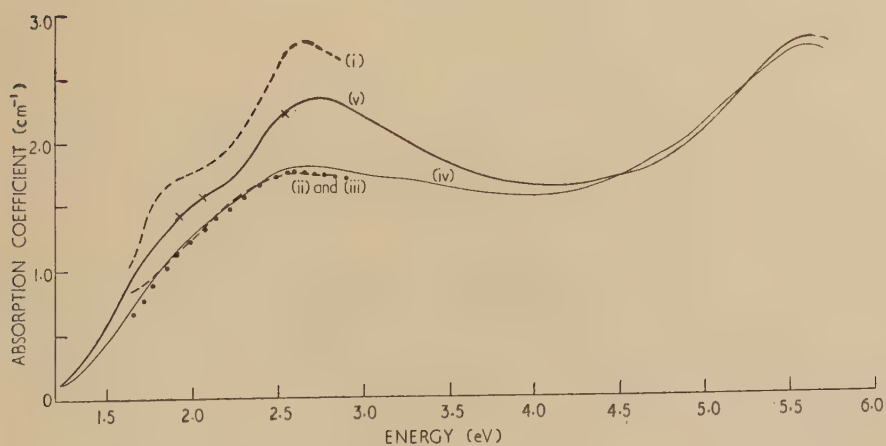
The absorption curves corresponding to (ii), (iii) and (iv) are in good agreement showing that the absorption of the centre is isotropic for any orientation of **E** perpendicular to the *c*-axis of the crystal. When the incident light is perpendicular to the *c*-axis, as in (ii) and (v), some ellipticity will be introduced but the effect is small, as may be seen from the transmission when the specimen is between crossed polaroids. Furthermore, the agreement between (ii) and either (iii) or (iv), in which only

Fig. 3



The anisotropic absorption in quartz at room temperature (full line) and at -195°C (broken line). The upper curves are type (i) and the lower type (ii) measurements.

Fig. 4



The anisotropic absorption of quartz at room temperature. The broken line refers to type (ii) and the dotted line to type (iii) measurements. The three crosses are the predicted type (v) results.

rotation is present, shows that $\mathbf{E}$ remains predominately in the initial direction. Using the curves for (i) and (ii) together with the polarization figure for the beam from the monochromator it is possible to predict the absorption coefficients for a type (v) measurement. Three points determined in this way are shown on fig. 4. It will be seen that they lie on the curve for the type (v) measurements.

Apart from those cases in which all types of measurement have been made on one specimen the ratio $\mu(\mathbf{E}_{\parallel}) : \mu(\mathbf{E}_{\perp})$ can be found from type (i) and (ii) measurements on a parallel cut specimen having one pair of polished surfaces. A summary of results is given in table 2.

Table 2

Specimen and irradiation	Position (ev) of absorption peak		$\mu\mathbf{E}_{\parallel}/\mu\mathbf{E}_{\perp}$ at 2.6 ev
	$\mathbf{E}_{\parallel}$	$\mathbf{E}_{\perp}$	
N.37 x-ray	2.8	2.7	1.45
N.36 γ -ray	2.71	2.60	1.54
N. 2 n°	2.75	2.60	1.56
N. 8 n°	2.70	2.60	1.51
N.14 n°	2.70	2.60	1.51
N.15 n°	2.70	2.60	1.54
N.35 n°	2.65	2.60	1.60
N.39 n°	2.75	2.63	1.55
N.40 n°	2.70	2.60	1.51

Although measurements have not been made on the C band (5.6 ev) with plane polarized light, the results of the type (iv) and (v) measurements shown in fig. 4 suggest that the absorption in this band is not significantly anisotropic.

§ 4. DISCUSSION

It is clear from these results that the A centres give rise to anisotropic absorption with respect to plane polarized light. Therefore it is necessary to know the crystallographic orientation and the instrumental polarizations when comparing the results from different specimens and from different kinds of irradiation, obtained in different laboratories.

A simple comparison may be made for type (iv) measurements since, in general, this will be independent of instrumental effects. Unfortunately the A_1 band is not observed in this case so that for a detailed comparison type (i) measurements must also be made. Since this involves using a parallel cut specimen type (ii) measurements can conveniently be made at the same time.

For the specimens which we have measured, the integrated absorption is greater when $\mathbf{E}$ is parallel to the c -axis than for any perpendicular direction. The concentration of centres involved is the same in the two

cases so that the transition probability for the centre must be anisotropic, being greater in the direction of the c -axis. An indication of the magnitude of this effect is given by the values of $\mu(\mathbf{E}||) : \mu(\mathbf{E}\perp)$ in table 2. Since the half-width does not vary significantly these values are equal, to within 5%, to the ratio of the sums of the f -values resolved in the two directions.

The Nature of the Centre

Brown and Thomas (1952) found that synthetic crystals coloured less readily than natural crystals and the results of Mitchell and Paige (1954) showed that the A_2 and A_1 bands were probably associated with impurity since the absorption did not increase with prolonged neutron irradiation. It was further shown by Ditchburn *et al.* (1954) that the effect was due to aluminium impurity.

The aluminium which, after ionization, is effective in producing the optical centre is probably incorporated substitutionally, as may be seen from the following experiments. Firstly, after heating for 12 hours in molten aluminium at 700°C a crystal became black throughout indicating that aluminium atoms have diffused for distances of the order of millimetres. Secondly, a non-uniformly coloured crystal, in which the visible absorption was in well-defined bands approximately parallel to a major rhombohedral face, was heated at 950°C for 24 hours and slowly cooled. After a second irradiation it was found that these bands were in the same positions in the crystal as before the heat treatment. The simplest interpretation of these experiments is that in the first we were dealing with diffusion via interstitial sites, while in the second the aluminium effective in producing colour centres was present substitutionally.

That the optical A bands and the paramagnetic resonance effects are associated with the same centre has been established by the magnetic measurements of Dr. Owen and Dr. Bagguley of the Clarendon Laboratory, Oxford, on two specimens in which the A_1 and A_2 bands have been completely removed. Dr. Owen has shown that the aluminium type of resonance was absent in a specimen in which the A bands had been removed by optical bleaching, and Dr. Bagguley found the same result on a specimen which had been radiation-bleached using a large neutron dose (Mitchell and Paige 1954).

It is concluded, therefore, that both optical and magnetic effects are associated with substitutional aluminium. This is in agreement with the model which has been proposed by O'Brien and Pryce to account for the magnetic results. In their model the substituted Al is assumed to be Al^- so that in the unirradiated state the binding is normal.* On irradiation an electron is released from the defect and is trapped either at some other impurity (O'Brien and Pryce 1954) or at a defect of the kind which can be produced by neutron damage (Ditchburn *et al.* 1954).

* It may be that the small differences in lattice parameter of different quartzes observed by Keith (1950) are due to varying concentrations of substitutional Al.

Wherever the electron is trapped O'Brien and Pryce suggest that the hole spends most of its time on an oxygen adjacent to the Al^- . The possible optical transitions on this model, using the O'Brien and Pryce nomenclature, are sp_z to p_x , or if the non-bonding sp_z is not filled between the bonding and non-bonding sp_z orbitals, or between p_y and non-bonding sp_z .

The origin of the line broadening is not understood, especially as the width is not greatly temperature dependent (Ditchburn *et al.* 1954). However, the anisotropy of the integrated absorption due to the centre must be associated with the directional property of the p_x , p_y or p_z orbitals since the transition is only allowed by virtue of the s-component of the other level. In the discussion of the wave functions given by O'Brien and Pryce, p_x is the component of the p wave function in a direction perpendicular to the planes of the Al^- - O^+ -Si triangles, p_y is the component parallel to the line joining Al^- and Si, while p_z is the component perpendicular to the line joining Al^- and Si. One bond in each of these triangles is at $44^\circ 15'$ to the c -axis and the other at $66^\circ 15'$. Using the data given by Wyckoff (1948) it is found that the normals to the six possible triangles make angles of $51^\circ 3'$ to the c -axis and the z directions $58^\circ 40'$, corresponding to the 54° between the Al^- -Si directions (y) and the c -axis.

We have to compare the measured f -value ratio with $\Sigma f_{\parallel}/\Sigma f_{\perp}$. In the parallel direction there are six components of dipole moment $M \cos \theta$ so that $\Sigma f_{\parallel} = C \cdot 6M^2 \cos^2 \theta$, where C is a constant. In the perpendicular direction, if $\mathbf{E}$ makes an angle ϕ with the projection in the basal plane of the required direction the components of dipole moment are two of $M \sin \theta \cos \phi$, two of $M \sin \theta \cos (\phi + 60)$ and two of $M \sin \theta \cos (60 - \phi)$. Then

$$\Sigma f_{\perp} = C \cdot 2M^2 \sin^2 \theta \{ \cos^2 \phi + \cos^2 (60 + \phi) + \cos^2 (60 - \phi) \},$$

which reduces to $C \cdot 3M^2 \sin^2 \theta$, being independent of ϕ . This agrees with the experimental result that the absorption was isotropic for all directions of $\mathbf{E}$ perpendicular to the c -axis.

The ratio of the integrated f -values, $\Sigma f_{\parallel}/\Sigma f_{\perp}$, is $2 \cot^2 \theta$ giving 1.3 when θ is $51^\circ 3'$, 1.06 for 54° and 0.74 for $58^\circ 40'$. The measured value is close to 1.5 (see table 2) so that we reject the possibility that the sp_z is the higher of the non-bonding orbitals. The agreement with the model if p_x is uppermost is considered reasonable. In order to obtain 1.5 from the model θ would have to be $49^\circ 6'$ which is only about 2° away from the normals to the triangles as calculated from the crystallography of undistorted quartz. Random variations about the $51^\circ 3'$ direction would lead to a higher value than 1.3 since $d/d\theta (\cot^2 \theta)$ decreases with increasing θ .

The anisotropic optical absorption of the A_1 and A_2 bands is therefore in reasonable agreement with the O'Brien and Pryce model if the p_x level is uppermost. However, the origin of the line broadening and the nature of the corresponding electron trap remain uncertain.

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CXLVII. CORRESPONDENCE

The Mean Lifetime and Some Properties of θ^0 -Particles : A Correction to Previously Published Data

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THE Manchester cloud chamber group, led by J. A. Newth, have recently pointed out a numerical error in table 2 of a paper by the author (Gayther 1954) on the mean lifetime of the θ^0 -particle. The value quoted for ϵ^* is 0.41, whereas the correct value is 0.82. This error does not affect the classification of the V^0 -decays described in the paper but it does affect the values of the momenta of the θ^0 -particles and thus their mean lifetime. The corrected values of the various quantities depending on ϵ^* are given in table 1. The explanation of the symbols is given in Gayther (1954).

Table 1. Corrected Data on 8 θ^0 -particles and 2 Probable θ^0 -particles

(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)
Event	$\gamma\beta$	P (MeV/c)	E (MeV)	W	t	T	T_1	T_2
					10 ⁻¹⁰ sec			
A1208	0.68	330	100	2.2	0.6	4.0	—	—
B1843	1.20	590	270	2.5	0.05	3.0	—	—
B1990	1.10	550	240	1.8	0.5	4.3	—	—
D1967	1.30	640	310	1.6	1.0	4.3	2.3	3.2
D3587	~7.0	~3400	~3000	4.5	0.1	0.5	—	—
D3754	0.73	360	120	3.8	0.6	6.8	3.7	4.8
D5123	1.30	640	310	1.7	0.7	3.7	2.0	2.8
D5155	2.50	1200	840	1.7	0.2	1.9	1.0	1.5
B3602	—	~780	~430	5.1	—	—	—	—
D319	—	~710	~370	1.7	—	—	—	—

The value of the mean life, τ , for the θ^0 -particle, found from the measurements on the first eight cases is given by

$$1/\tau = (1.8 \pm 0.8) \times 10^{10} \text{ sec}^{-1}$$

$$\text{i.e. } \tau = (0.6^{+0.4}_{-0.2}) \times 10^{-10} \text{ sec.}$$

† Now at A.E.R.E., Harwell.

This corrected result may be combined with the data of other groups listed in table 6 of Gayther (1954) to obtain a weighted mean value (from 34 events) of

$$\tau = (1.7^{+0.7}_{-0.4}) \times 10^{-10} \text{ sec.}$$

In a recent paper Gayther and Butler (1955)* made further use of the data on θ^0 -particles given by Gayther (1954). Owing to the error in the original value of ϵ^* the values of the momenta and kinetic energies of the θ^0 -particles given in table 1 of the G.B. paper are incorrect, thus all the conclusions on the production processes of θ^0 -particles need modification. The corrected value of the momenta and energies are given above in columns (3) and (4) of table 1. Column (5) contains the corrected weighting factors, W ; the derivation and significance of these factors are described in § (3) of G.B.

The differential kinetic-energy spectrum and angular distribution of Λ^0 -particles were discussed by G.B. The behaviour of these particles is similar to that of the 'grey' protons from stars. The corrected data on the differential energy spectrum for θ^0 -particles is given in table 2, where a comparison is made with Λ^0 -particles. The shape of the angular distribution for the θ^0 -particles given in fig. 3 of G.B. is not significantly affected by the change in the weighting factors.

Table 2. The Differential Energy Spectra for θ^0 - and Λ^0 -particles

Kinetic energy	<50 MeV	50-100 MeV	100-400 MeV	400-700 MeV	700-1000 MeV	>1000 MeV
Corrected no. of θ^0 -particles .. (actual no. ob- served 10)	—	—	15	5	2	4
Corrected no. of Λ^0 -particles .. (actual no. ob- served 28)	28	10	26	8	—	—

Superficially the θ^0 -particles appear to be of higher energy than the Λ^0 -particles. It is necessary, however, to consider the bias effects at low energy due to the size of the cloud chamber and the short lifetime of the particles. Reference to fig. 1 of the G.B. paper shows that for Λ^0 -particles in the energy range (50-100) mev the detection probability ranges from 0.45 to 0.50. For θ^0 -particles in the same energy interval the probability range is 0.30 to 0.42. In this particular energy interval a corrected number of 10 Λ^0 were observed (actual events 5) and *no* case of a θ^0 . Thus there is some evidence that the differential energy spectrum of θ^0 -particles probably falls below 100 mev, while Gayther and

* Subsequently referred to as G.B.

Butler concluded that for Λ^0 -particles it rises. There is some provisional evidence that the spectrum of the θ^0 -particles is similar to that of the $\pi^\pm$ -mesons produced in stars which, according to Camerini *et al.* (1950), probably falls for energies of less than 100 mev.

The analysis of Gayther and Butler dealt with a total of 45 V^0 -particles, 28 were identified as Λ^0 -particles, while 10 were θ^0 -particles; the remaining 7 cases were unidentified. The relative number of Λ^0 - and θ^0 -particles at production can be determined using the detection probabilities W . The corrected data give

$$N_{\Lambda^0} : N_{\theta^0} \equiv (90 \pm 17) : (27 \pm 9)$$

assuming that all the unidentified events were due to Λ^0 -particles. If, however, it is assumed they were θ^0 -particles, then

$$N_{\Lambda^0} : N_{\theta^0} \equiv (72 \pm 16) : (44 \pm 12).$$

Taking into account the possibility of detection bias effects discussed in G.B., it now seems that more Λ^0 - than θ^0 -particles are produced in nuclear interactions in lead.

A method for finding the frequency of production of Λ^0 - and θ^0 -particles with respect to the number of shower particles was described by G.B.; the corrected results are

$$N_{\Lambda^0}/N_{\pi^\pm} \simeq 3\% ; N_{\theta^0}/N_{\pi^\pm} \simeq 1.3\%.$$

The data refer to V -decays in showers containing less than 9 shower particles.

In conclusion it may be pointed out that if more Λ^0 - than θ^0 -particles are produced in energetic nuclear interactions, there must be processes other than the double production process.

$$\pi^- + p \rightarrow \Lambda^0 + \theta^0,$$

first reported by W. B. Fowler *et al.* (1953). One possible additional source of Λ^0 -particles may be the nuclear capture of K^- -particles. Barker (1953) and De Staebler (1954) have reported evidence for the processes

$$K^- + p \rightarrow \Lambda^0 + \pi^0$$

$$\text{and } K^- + n \rightarrow \Lambda^0 + \pi^-.$$

If the K^- -particles are readily captured in this manner, a considerable number of Λ^0 may be produced unassociated with θ^0 -particles.

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CXLVIII. REVIEWS OF BOOKS

Advanced Mathematics for Engineers. By H. W. REDDICK and F. H. MILLER. [Pp. xiv+548.] (New York: John Wiley and Sons, Inc.; London: Chapman and Hall, Ltd.) (3rd edition, 1955.) Price 52s.

THE third edition of this excellent text-book can be warmly welcomed. The range of topics is extremely wide and physical applications of the basic mathematics are well illustrated by problems worked in the text. In addition, lists of problems are provided for the student to try, with answers given at the end of the book. For the third edition, these lists have been extensively revised and the total number of problems has been increased. E. N. F.

An Introduction to Stochastic Processes, with Special Reference to Methods and Applications. By M. S. BARTLETT. [Pp. xiv+312.] (Cambridge: University Press.) Price 35s.

THIS long-awaited book is most welcome. Statisticians have been familiar for some time with Professor Bartlett's North Carolina lecture notes; the present work has developed out of these notes into an epoch-making general treatise which concentrates on describing and applying stochastic processes: the basic mathematical theory, while touched on, is left to be developed more fully by Mr. J. E. Moyal in a companion volume which should be published before long.

The older generation of statisticians, in this country at least, paid little attention before the war to the proper methods of dealing with data for which independence of individual elements could not be assumed: for example, analysis of economic data could hardly be regarded as satisfactory when done by stock methods. That generation, because of the age factor, is having difficulty in assimilating the new knowledge in its present rapid state of growth, and so will welcome a book which summarizes progress to date: the book, of course, is a 'must' for the younger generation of science students. The greater part of it takes us through what must be regarded as a necessary standard treatment of the various aspects of stochastic processes. Chapter 5 summarizes some of the more formal mathematics of limiting stochastic operations, although a full treatment is left to Moyal, while towards the end the basic elements of 'classical' statistical analysis are applied to the processes, with particular reference to the analysis of time series.

It is impossible in a short notice to give a proper summary of the main part of the book, but mention of the problems dealt with in various scientific fields will be a sufficient indication of its all-embracing character. In physics the author deals with crystal lattices, diffusion, cascade showers, molecular association in statistical mechanics, and the fluctuation theory of molecular concentrations, with an aside on the Monte Carlo method. In engineering the important theory of communication gets a large share of attention. Genetics is represented by the study of genetic recombination, the theory of inbreeding, natural selection and population genetics generally. There are important population problems in regard to fluctuations, and birth and death processes; in biology growth and mutation in bacterial populations are studied, likewise contagious distributions; in the medical field the theory is applied to epidemics. More generally, there are industrial applications in questions such as the renewal of articles with effective random lifetimes, sequential analysis, and the theory of queues, as applied to the servicing of machines and control of aircraft landings. Examples are given of insurance risks, of the response of a learner to a stimulus, and of the varying fertility of an area of land. This will be enough to show that the book will be of interest to a much wider class of persons than the mathematical statisticians, and the author is to be congratulated on his industry. J. W.

Chemistry of the Solid State. Edited by W. E. GARNER. [Pp. viii+417.] (London: Butterworths Scientific Publications; New York: Academic Press Incorporated.) 50s.

THIS work is a collection of essays, many of them by Professor Garner, his staff and former pupils. It would be better described as the chemistry of reaction in the solid state, dealing as it does with nucleation, photochemical effects and so on. Several of the articles describe the work carried out in Professor Garner's own laboratory. It is a useful and stimulating volume, and the reviewer's chief feeling on looking through it was that some of the articles ought to be expanded into a book. Or alternatively that this new subject, the chemistry of reactions in the solid state, needs a new book that would attempt a synthesis and compare what is known about the compounds discussed with similar information about metals. Perhaps one of the authors of this volume will attempt it. They are:

- F. C. Frank—Chemistry of Crystal Dislocations ;
- F. S. Stone—Lattice Defects in Ionic Crystals ;
- P. W. M. Jacobs and F. C. Tompkins—Action of Light on Solids ;
- P. W. M. Jacobs and F. C. Tompkins—Surfaces of Solids ;
- T. J. Gray—Semi-conductivity and Magneto-chemistry of the Solid State ;
- W. J. Dunning—Theory of Crystal Nucleation from Vapour, Liquid and Solid Systems ;
- P. W. M. Jacobs and F. C. Tompkins—Classification and Theory of Solid Reactions ;
- W. E. Garner—Kinetics of Endothermic Solid Reactions ;
- W. E. Garner and L. L. Bircumshaw—Kinetics of Exothermic Solid Reactions ;
- C. E. H. Bawn—Decomposition of Organic Solids ;
- A. R. Ubbelohde—Explosion and Detonation in Solids ;
- A. J. E. Welch—Solid-Solid Reactions ;
- J. W. Mitchell—Photographic Process ;
- T. B. Grimley—Oxidation of Metals ; and
- F. S. Stone—Electronic Factor in Chemisorption and Catalysis.

N. F. M.

[The Editors do not hold themselves responsible for the views expressed by their correspondents.]

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